

# THE PROCEEDINGS OF THE PHYSICAL SOCIETY

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No. 273

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# THE PROCEEDINGS OF THE PHYSICAL SOCIETY

VOL. 49, PART 4

JULY 1, 1937

No. 273

## THE SATURATION-DISCRIMINATION OF TWO TRICHROMATS

By W. D. WRIGHT, D.Sc., A.R.C.S., D.I.C.

AND

F. H. G. PITT, Ph.D., A.R.C.S., D.I.C.,

Imperial College of Science and Technology

*Received 21 October 1936. Read 28 May 1937*

**ABSTRACT.** Measurements have been made by the authors to determine the values for their own eyes of  $\log \{(L_W + L_S)/L_S\}$  for the just-noticeable first step from white for a range of spectral colours.

THIS short paper is supplementary to one<sup>(1)</sup> published in 1935, in which the detailed characteristics of the authors' colour-vision were tabulated. We expressed some dissatisfaction with the data on saturation-discrimination, not because we doubted the accuracy of the measurements but because, when the first step from the spectral locus was measured, each observation was necessarily made in a different part of the colour field, so that the sensitivity of the eye to colour-differences was itself variable. The results thus represented two variables, namely the sensitivity of the eye and the colour valency of the spectral colours, whereas the latter alone should have been measured. It can be effectively isolated if the first step from white is measured rather than the first step from the spectral locus, since the observations will all be made in practically the same part of the colour triangle.

No very simple way of making the measurements with the Wright colorimeter was available at the time, and the less satisfactory step from the spectral locus was measured instead. A straightforward adaptation of the colorimeter has since been devised and the modification and method of observation have been described by Nelson<sup>(2)</sup>. The new data obtained in this way for the authors' eyes are shown in figures 1 and 2. The saturation-discrimination has been represented in the conventional manner as  $\log \{(L_W + L_S)/L_S\}$ , where  $L_S$  is the luminosity of the spectral colour that has been mixed with white of luminosity  $L_W$  to produce the just-noticeable step. A radiation with a small colour valency will thus give a small value of



$\log \{(L_W + L_S)/L_S\}$ , and *vice versa*. The steps were measured relative to the white given by the C.I.E. 1931  $S_B$  source, which has a colour temperature of  $4800^\circ \text{K}$ .

The general agreement between the authors' curves and the mean curve given

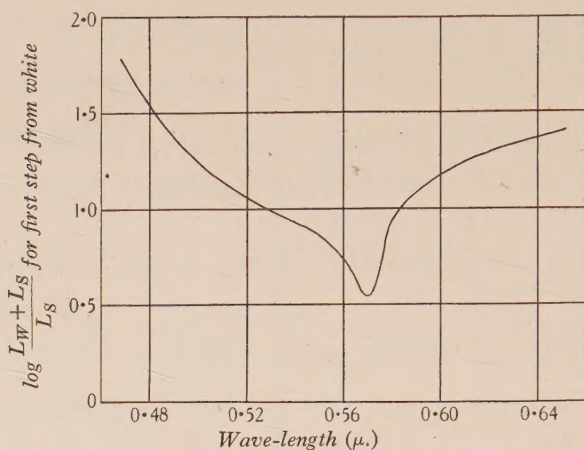


Figure 1. Saturation-discrimination curve for F. H. G. P.

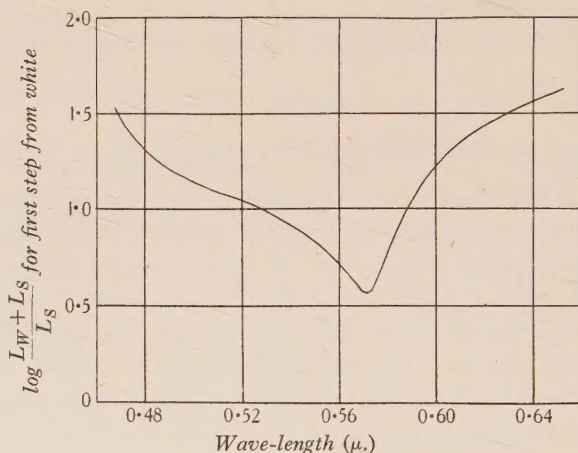


Figure 2. Saturation-discrimination curve for W. D. W.

by Martin, Warburton and Morgan<sup>(3)</sup> is very good. Undoubtedly the most striking feature is the pronounced minimum shown in all the curves; the very close agreement between one observer and another in the wave-length, about  $0.57 \mu.$ , at which the minimum occurs, is specially noteworthy.

## Saturation-discrimination for F. H. G. P. and W. D. W.

Wave-length $\lambda$ ( $\mu$ .)	Saturation-discrimination, $\log \frac{L_W + L_S}{L_S}$		Wave-length $\lambda$ ( $\mu$ .)	Saturation-discrimination, $\log \frac{L_W + L_S}{L_S}$	
	F. H. G. P.	W. D. W.		F. H. G. P.	W. D. W.
0.47	1.77	1.48	0.57	0.55	0.58
0.48	1.55	1.32	0.58	0.92	0.75
0.49	1.40	1.22	0.59	1.09	1.02
0.50	1.26	1.15	0.60	1.18	1.21
0.51	1.14	1.09	0.61	1.25	1.35
0.52	1.05	1.04	0.62	1.29	1.43
0.53	0.98	0.98	0.63	1.34	1.51
0.54	0.93	0.92	0.64	1.36	1.57
0.55	0.86	0.83	0.65	1.40	1.62
0.56	0.73	0.72			

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- (2) NELSON, J. H., page 332 of this volume.
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## THE COLOUR-VISION CHARACTERISTICS OF A TRICHROMAT, PART 2

BY J. H. NELSON, A.R.C.S., D.I.C., B.Sc.

*Received 21 October 1936. Read 28 May 1937*

**ABSTRACT.** The author's luminosity curve, trichromatic coefficients, mixture curves, complementary wave-lengths and their mixture in terms of luminosity, and the hue- and saturation-discrimination curves have been obtained and the results tabulated.

IN a recent paper by Wright and Pitt<sup>(1)</sup> the importance of having the complete characteristics of individual observers was pointed out, and they gave the data for their own eyes. In this paper the characteristics of the present author, also a trichromat, are given. The apparatus used for the measurements was the Wright colorimeter, which has been fully described elsewhere<sup>(2)</sup>.

The methods of expressing the results have been dealt with previously, and it is only necessary to repeat them very briefly. The trichromatic coefficients are expressed on the W. D. W. system, in which the primaries are red  $0.65 \mu$ ., green  $0.53 \mu$ . and blue  $0.46 \mu$ ., and the units are based on two monochromatic matches at  $0.5825 \mu$ . and  $0.4940 \mu$ . The luminosity is referred to an equal-energy spectrum, the maximum being taken as unity.

The position of the maximum of the luminosity curve for the observer J. H. N. is  $0.552 \mu$ ., which compares with  $0.558 \mu$ . for W. D. W.,  $0.562 \mu$ . for F. H. G. P., and  $0.555 \mu$ . for the standard observer. The position of the standard illuminant B in the colour triangle for the observer J. H. N. lies farther towards the yellow than the normal; this is the reverse of what holds for the observers W. D. W. and F. H. G. P. This indicates a slightly more dense macular pigment than the normal, a conclusion which is further supported by the luminosity curve, which shows a lower luminosity in the blue than those of either W. D. W., F. H. G. P. or the normal.

The complementary wave-lengths have been calculated from the colour triangle, together with the proportions, in luminosity, in which they should be mixed to give the standard illuminant B.

The hue-discrimination for the observer J. H. N. is similar to that for the previous two observers, showing a secondary minimum in the violet, and not in the red.

As has been pointed out by Wright and Pitt, since the saturation-discrimination does not vary very much when measured from the spectral locus, it would be instructive to know the variation of the saturation-discrimination when measured from the white point; the requisite data were therefore obtained by adding a small



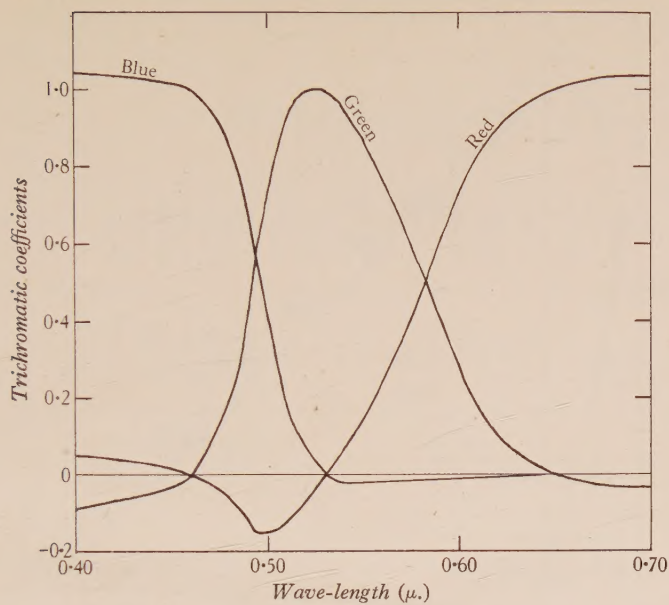


Figure 1. Spectral coefficients.

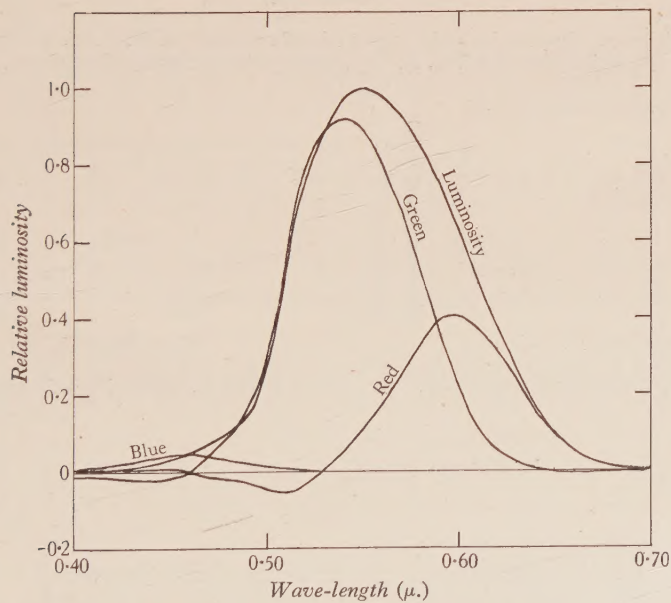


Figure 2. Equal-energy luminosity and spectral mixture curves.

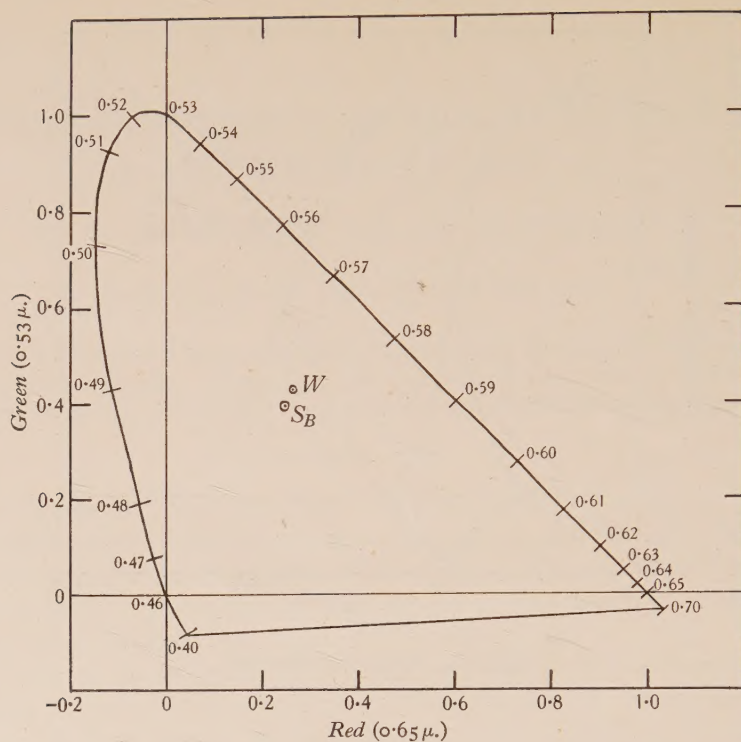


Figure 3. Spectral locus in colour triangle plotted on W. D. W. system.  
 $W$ , white point for J. H. N.  $S_B$ , white point for standard observer.

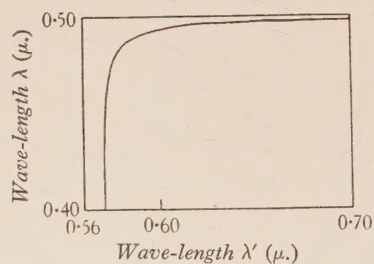


Figure 4. Complementary wave-lengths.

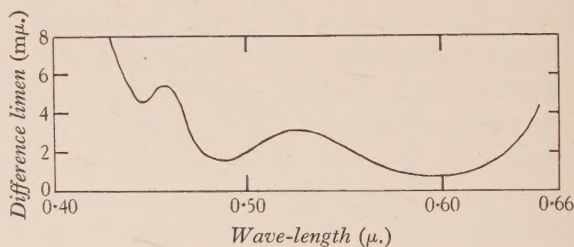


Figure 5. Hue-discrimination.

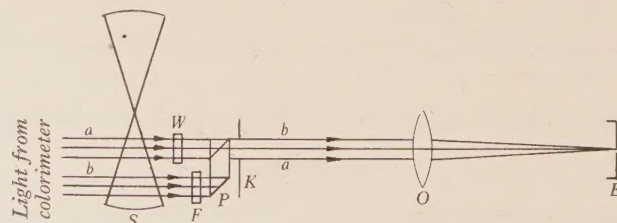


Figure 6. Diagram of optical system for saturation-discrimination.



Wave-length $\lambda$ ( $\mu$ )	Relative luminosity of equal- energy spectrum	Trichromatic coefficients			Mixture curves for equal- energy spectrum			Complementary wave-lengths ( $\mu$ )		Lumino- sity of $\lambda$ to be mixed with unit luminosity of $\lambda'$	Hue-dis- crimination; just- noticeable wave- length differences ( $\mu$ )	Saturation- discrimination, $\log \frac{L_W + L_S}{L_S}$	Wave-length $\lambda$ ( $\mu$ )
		B (0.46 $\mu$ )	G (0.53 $\mu$ )	R (0.65 $\mu$ )	$L_B$	$L_G$	$L_R$	$\lambda$	$\lambda'$				
0.40	0.0007	1.043	-0.093	0.050	0.0052	-0.0095	0.0038	—	—	—	—	—	0.40
0.41	0.0028	1.038	-0.081	0.043	0.0098	-0.0113	0.0043	0.41	0.5710	0.01006	—	—	0.41
0.42	0.0070	1.033	-0.069	0.036	0.0165	-0.0156	0.0061	0.42	0.5712	0.0132	—	—	0.42
0.43	0.0125	1.028	-0.057	0.029	0.025	-0.020	0.0075	0.43	0.5715	0.0164	0.0080	—	0.43
0.44	0.0196	1.023	-0.046	0.023	0.0325	-0.0202	0.0073	0.44	0.5718	0.0195	0.0054	—	0.44
0.45	0.0270	1.018	-0.035	0.017	0.0404	-0.0204	0.007	0.45	0.5722	0.0227	0.0048	—	0.45
0.46	0.0468	1.000	0.000	0.000	0.0468	0.000	0.000	0.46	0.5729	0.0345	0.0054	—	0.46
0.47	0.075	0.951	0.076	-0.027	0.039	0.051	-0.015	0.47	0.5750	0.0676	0.0032	1.125	0.47
0.48	0.109	0.862	0.194	-0.056	0.030	0.099	-0.020	0.48	0.5780	0.137	0.0018	1.08	0.48
0.49	0.150	0.683	0.430	-0.113	0.018	0.162	-0.031	0.49	0.5894	0.386	0.0016	1.04	0.49
0.50	0.274	0.412	0.736	-0.148	0.012	0.306	-0.044	—	—	—	0.0021	1.04	0.50
0.51	0.528	0.190	0.930	-0.121	0.008	0.575	-0.055	—	—	—	0.0027	0.995	0.51
0.52	0.763	0.070	0.998	-0.068	0.004	0.797	-0.038	—	—	—	0.0031	0.96	0.52
0.53	0.890	0.000	1.000	0.000	0.000	0.890	0.000	—	—	—	0.00315	0.916	0.53
0.54	0.973	-0.020	0.945	0.075	0.000	0.921	0.052	—	—	—	0.0029	0.85	0.54
0.55	0.998	-0.019	0.868	0.151	0.000	0.888	0.110	—	—	—	0.0024	0.78	0.55
0.56	0.978	-0.018	0.776	0.242	0.000	0.801	0.177	—	—	—	0.0018	0.65	0.56
0.57	0.928	-0.014	0.664	0.350	0.000	0.677	0.249	—	—	—	0.00135	0.475	0.57
0.58	0.850	-0.008	0.534	0.474	0.000	0.515	0.335	0.4838	0.58	0.204	0.00105	0.76	0.58
0.59	0.753	-0.007	0.404	0.603	0.000	0.356	0.397	0.4909	0.59	0.428	0.0009	0.93	0.59
0.60	0.625	-0.007	0.277	0.730	0.000	0.221	0.404	0.4940	0.60	0.707	0.0008	1.03	0.60
0.61	0.491	-0.006	0.178	0.828	0.000	0.114	0.377	0.4958	0.61	0.983	0.0009	1.150	0.61
0.62	0.371	-0.006	0.101	0.905	0.000	0.051	0.320	0.4967	0.62	1.18	0.0012	1.245	0.62
0.63	0.261	-0.004	0.052	0.952	0.000	0.019	0.242	0.4973	0.63	1.33	0.0018	1.33	0.63
0.64	0.173	-0.002	0.022	0.980	0.000	0.005	0.168	0.4975	0.64	1.42	0.0028	1.435	0.64
0.65	0.094	0.000	0.000	1.000	0.000	0.000	0.094	0.4976	0.65	1.51	0.0044	1.485	0.65
0.66	0.048	0.000	-0.022	1.022	0.000	-0.001	0.049	—	—	—	—	1.54	0.66
0.67	0.023	0.000	-0.030	1.030	0.000	-0.009	0.0239	—	—	—	—	1.62	0.67
0.68	0.013	0.000	-0.031	1.031	0.000	-0.008	0.0138	—	—	—	—	1.66	0.68
0.69	0.0085	0.000	-0.032	1.032	0.000	-0.0005	0.0090	—	—	—	—	1.72	0.69
0.70	0.0059	0.000	-0.033	1.033	0.000	-0.00028	0.00618	0.4979	0.70	1.62	—	1.75	0.70
1931 C.I.E. source $S_B$		0.307	0.431	0.262	Relative luminosities of trichromatic units of primaries								
$\lambda = 0.4940\mu$		0.572	0.572	-0.144	0.46 $\mu$	0.53 $\mu$	0.65 $\mu$						
$\lambda = 0.5825\mu$		-0.008	0.504	0.504	0.68	1.000	0.711						

quantity of spectral colour to white (standard illuminant B). For this purpose it was necessary to make some additions to the apparatus, the optical system necessary in addition to the colorimeter being shown in figure 6. A brass sector *S*, coated with magnesium oxide, is illuminated by the light from a lamp as specified by the C.I.E. (1931) for standard illuminant B<sup>(3)</sup>. The sector is arranged so that both halves of the photometric field in the prism *P* are filled with white light during one half of the revolution, and that during the other half of the revolution a monochromatic radiation of known wave-length is admitted to the top half of the field, forming beam *b*. This monochromatic radiation is obtained from the spectrum supplying the instrument primaries, and the intensity is controlled by a calibrated photometer wedge. The intensity of the white in the top half of the field (beam *b*) may be controlled by neutral filters *F*, but it is advisable to use only the lightest filters as the darker ones

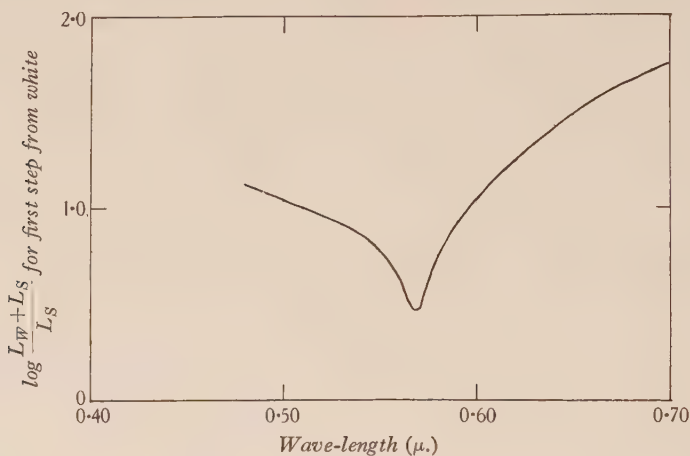


Figure 7. Saturation-discrimination.

may deviate somewhat from neutrality. In the lower half of the field the intensity of the white forming beam *a* is controlled by a very shallow photometer wedge. The sector is arranged to run at two speeds. The fast speed is fast enough to remove flicker and thus provide two uniform fields, a white *a* and a white-plus-monochromatic *b*, between which a saturation step may be made. At the slow speed the intensities of the white and the monochromatic radiation can be compared by means of flicker.

When the observations are being taken the sector is first run slowly, and the observer is asked to equate the intensities of the white and the monochromatic radiation by adjusting the intensity of the latter so that flicker may be removed. The sector is then run fast, and the observer is asked to adjust the intensity of the monochromatic radiation until he can just notice a difference in colour between the top and bottom halves of the field, the intensities of both halves being kept the same by adjusting the intensity of the white of the bottom half of the field. In this way the intensity of monochromatic radiation to be added to the white to make one step



can be found. If  $L_W$  is the intensity of the white and  $L_S$  that of the added monochromatic radiation, then the saturation step can be expressed as the logarithm of the ratio  $(L_W + L_S)/L_S$ . When this fraction is large, only a small quantity of monochromatic radiation is needed to cause a just noticeable change in colour.

The measurements of saturation-discrimination taken by this method may be compared with the results obtained by Martin, Warburton and Morgan<sup>(4)</sup>. The curve given in the present paper falls below that given by them in the blue-green region, but its general shape is the same.

The author wishes to thank Dr W. D. Wright for his help, and the Medical Research Council for their financial assistance.

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#### DISCUSSION

Mr J. GUILD. Papers of this kind, giving full details of the colour-vision of individual observers, are of great value. One of the main difficulties in correlating the data on colour-vision accumulated in the past has been due to the fact that the various functions—visibility, colour-matching, hue-discrimination, etc.—have usually been determined by different people using different apparatus and standards. From the point of view of providing knowledge of the visual processes it is much more important to have comprehensive information about a few people than to have a little information about many people. If the programme followed at the Imperial College provides us with one or two more individual studies of this kind it will be of great value. I note that the evidence of this paper and of the former paper by Wright and Pitt goes to show that the wave-length of maximum visibility is on that side of the value  $0.555 \mu$ . of the C.I.E. standard visibility function which corresponds to the departure from normality in macular pigmentation, as is indicated by the position of the white point on Dr Wright's form of the colour diagram. This confirms that the standard value must be fairly nearly correct for normal observers, despite the fact that this view has recently been called in question.

# THE EFFECT OF SECONDARY BETA RADIATION ON GAMMA-RAY MEASUREMENTS MADE IN AIR

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**ABSTRACT.** A sensitive method of measuring small ionization currents is described, and the effect of secondary beta radiation upon ionization measurements of gamma radiation is examined. It is shown that if ionization chambers made of light-atom materials have walls less than 4.0 mm. thick, secondary beta radiation is recorded so that readings are in excess of the true values. The results explain some discrepancies found by other workers.

## § 1. INTRODUCTION

FOR the evaluation of the gamma-ray dosage in experiments on the biological effects of gamma radiation it is often necessary to make ionization measurements in air at small distances from radium sources. The literature shows that measurements made in air in the vicinity of radium sources may give values that differ widely from what might have been expected. Spear and Grimmett<sup>(1)</sup> for example found that the intensity-values due to a one-gramme radium source were consistently 24 per cent higher than the values obtained by calculation. They measured the intensities in air at distances of from 3 to 8 cm. from the radium with condenser chambers of the Sievert pattern<sup>(2)</sup> which were constructed of elektron metal and had walls 1.0 mm. thick. Similarly Benner<sup>(3)</sup> found that for distances of from 3 to 14 mm. from a radium source, the intensity-values measured in air with ionization chambers having walls of elektron metal and brass respectively, of thickness 0.3 mm., were always considerably (14 per cent or more) in excess of those calculated.

The purpose of this paper is to describe some experiments which show that the discrepancies referred to can be ascribed to secondary beta radiation from the metal filters of the radium sources. If the chamber wall is sufficiently thin such radiation can enter the air volume of the chamber and add considerably to the ionization produced by gamma radiation. On the other hand, if the wall be thickened by the necessary amount no secondary beta radiation will be recorded.

## § 2. APPARATUS AND METHODS

The ionization chamber and current-measuring system that formed the main part of the apparatus are shown diagrammatically in figure 1. The ionization chamber was of the thimble type and made of elektron metal with a wall 0.5 mm.



thick. Its air volume was  $1.31 \text{ cm}^3$ . The lead from the graphite inner electrode of the chamber is carried to the Lindemann electrometer on a number of polished amber buttons through an air-tight brass tube that is continuously evacuated to avoid ionization losses. The tube also serves as an earthed screen for the lead.

For measuring ionization currents a null method is used, the potential across the capacity  $C$  being continuously increased to maintain the needle of the instrument at earth potential. The processes of isolating and earthing the electrometer needle start and stop the stop watch incorporated in the circuit, figure 1, so that the time taken for a measured change of potential is automatically recorded. If ionization

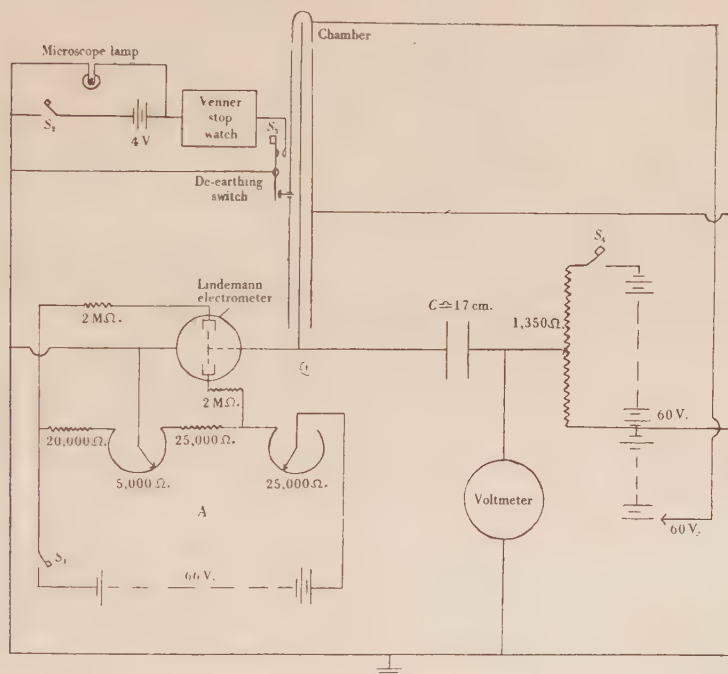


Figure 1. Arrangement for measuring ionization currents.

currents are to be compared, the potential-change per second is a suitable measure. If the capacity  $C$  is known an absolute determination of the current can be made. Thus with a graphite ionization chamber the apparatus serves as an X-ray dosimeter. This, in fact, is its routine use. The auxiliary circuit  $A$ , figure 1, is solely for the purpose of providing an easy control upon the sensitivity and zero setting of the electrometer<sup>(4)</sup>.

The construction and mounting of the ionization chamber is shown in figure 2, which also illustrates how the radiation source was set up relative to the chamber. The lead to the electrode is carried through an amber plug which is highly polished and waxed into the brass tube to form an air-tight joint. The thimble fits tightly over the shoulder on the amber plug. The source of gamma rays consists of radon in glass capillaries which are contained in a platinum tube with a wall 0.5 mm. thick.

The latter is sufficient to absorb all primary beta radiation. In all the experiments the source and ionization chamber were arranged parallel to one another and the source was mounted in a holder attached to the evacuated lead. Thus, if any movement of the lead and its chamber occurred, the distance between the source and the chamber did not change.

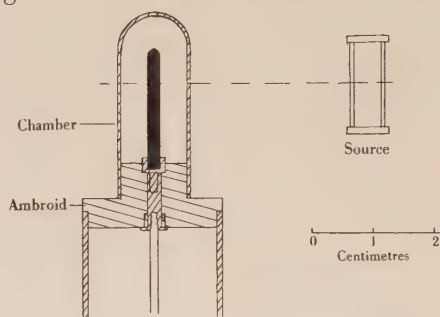


Figure 2. Dispositions of ionization chamber and radon source.

A series of sheaths of various thicknesses were made in elektron metal to fit tightly over the original chamber so that the wall-thickness of the chamber could be increased from 0.5 mm. up to 6.0 mm. as required.

### § 3. EXPERIMENTS

*The effect of large metal filters upon the ionization recorded in chambers with walls of various thicknesses.* For this experiment two circular metal discs were used as secondary filters. These were of copper and lead respectively. Each was 5.0 cm. in diameter and 0.5 mm. thick. When added to the arrangement shown in figure 2 they were placed between the source and the chamber so that the source was in contact with the centre of the filter. The distance between the centres of the source and chamber was 3.0 cm.

A series of measurements of the ionization current were made under three sets of conditions, (a) with no extra filter present, (b) with the copper filter added and (c) with the lead filter added. For each set of conditions readings were taken for different thicknesses of the wall of the chamber. Alternate records were taken in order to allow for decay of the radon source during the period of the experiment. Figure 3 shows the results expressed in graphical form.

It is seen that when the chamber wall is only 0.5 mm. thick the introduction of the metal filter causes the ionization to increase very markedly in spite of the absorption of gamma radiation that must occur. This increase is about 19 per cent for the copper sheet and as much as 32 per cent for the sheet of lead.

Two distinct types of results are obtained according to whether the large secondary filters are included or not. When no filter is added the ionization in the chamber increases with increase in wall-thickness up to a maximum value when the wall is about 2 mm. thick, after which a slow fall takes place. When either the copper



or lead filter is included, the ionization decreases continuously with increasing wall-thickness. Such a result as the latter is to be explained in terms of a strong secondary beta radiation from the filters, which augments the ionization for the smallest wall-thickness and is progressively absorbed as the wall-thickness is increased. Finally the results obtained with the filters present are lower than those with no filter, in consequence of absorption of the primary gamma rays in the filters. The preliminary rise in the curve expressing the results with no filter added is due to extra ionization by recoil electrons liberated from the thickened wall of the chamber. This question has received considerable attention from Mayneord and Roberts<sup>(5)</sup>.

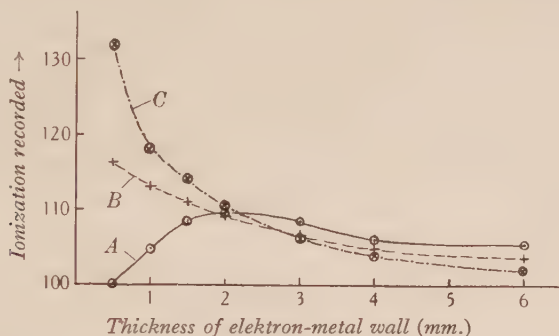


Figure 3. Showing how the current in an ionization chamber is affected by secondary radiation from a metal filter. *A*, no filter included; *B*, copper filter included; *C*, lead filter included.

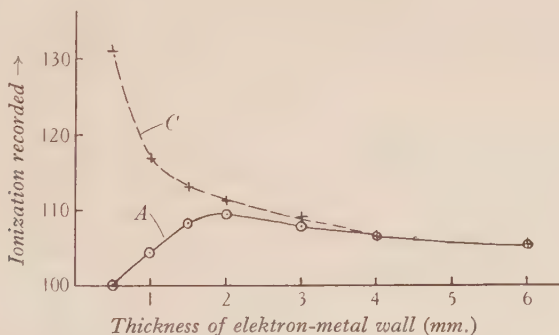


Figure 4. Showing the elimination of the effects of secondary beta radiation by increasing the wall-thickness of the ionization chamber. *A*, no filter included; *C*, lead filter included.

As is to be expected from the work of Stahel<sup>(6)</sup>, the amount of secondary beta radiation liberated from the lead sheet is very much greater than that from the copper sheet.

*The wall-thickness that absorbs all secondary beta radiations from metal filters.* In this experiment the same lead filter was used as before but a hole was cut at its centre having exactly the shape and size of the platinum tube that contained the radon source. Thus when the filter was placed in position as in the first experiment no absorption of primary gamma radiation occurred in it. The distance between the source and ionization chamber was 3.0 cm. as before.

Measurements were made of the ionization current within the chamber with and without the filter for the range of wall-thicknesses 0.5 mm. to 6.0 mm.; the results obtained are expressed graphically in figure 4. It is seen that when the chamber wall has a thickness of 4.0 mm. or more, the ionization recorded is not changed by the addition of the filter. Thus all secondary beta radiation is absorbed in the wall of the ionization chamber if its thickness is 4.0 mm. or more. Therefore any ionization chamber of light-atom material that is used for measuring gamma radiation in air is liable to record secondary beta radiation from nearby metal parts if its walls are not at least 4.0 mm. thick.

It seems then that the excess values found by Spear and Grimm<sup>(1)</sup> were due to the fact that their ionization chambers had elektron metal walls only 1.0 mm. thick, while a large disc of brass was used as the primary radium filter. To confirm this result a further experiment was carried out.

*Comparison of readings made with thin-walled and thick-walled ionization chambers when the large brass disc used by Spear and Grimm<sup>(1)</sup> is present.* The actual brass filter, 10 cm. in diameter and 2.3 mm. thick, used by Spear and Grimm<sup>(1)</sup> was available, and an aperture was cut in it in exactly the same manner as in the case of the lead sheet used in the second experiment. It was therefore possible to add this in the same way to the arrangement already described (figure 2) without absorbing any primary gamma radiation.

Readings were made of the ionization currents with and without the brass disc in position, an elektron-metal ionization chamber with walls (a) 1.0 mm. thick and (b) 4.0 mm. thick being used. The results are summarized in the table for various distances between the source and chamber, in terms of the ratio of the readings obtained with and without the disc for the two wall-thicknesses.

Table. Comparison between readings obtained with thin-walled and thick-walled ionization chambers

Distance between source and ionization chamber	Ionization with brass disc ionization with no brass disc	
	1.0-mm. wall	4.0-mm. wall
2.24 cm.	1.083	1.000
3.00 "	1.105	1.012
3.96 "	1.130	0.985
5.08 "	1.140	1.000
6.03 "	1.162	1.008
7.00 "	1.144	1.004

These results show that when the wall of the ionization chamber is only 1.0 mm. thick, the brass disc adds from 8 to 16 per cent to the ionization recorded, whereas if the wall is 4.0 mm. thick the presence of the brass does not affect the result within the experimental error. In the experiments of Spear and Grimm<sup>(1)</sup> the disc would serve as an even more efficient radiator, since the radium was spaced round a circle 8.0 cm. in diameter so that more of the metal surface was exposed to its action.



#### § 4. DISCUSSION OF RESULTS

If satisfactory measurements of gamma radiation are to be made in air with ionization chambers of light-atom materials in the vicinity of radium sources, the walls of the chamber must be at least 4.0 mm. thick in order to avoid excess ionization by secondary beta radiation. It would therefore appear to be impracticable to make reliable measurements of gamma radiation at distances less than 4.0 mm. from a radium source. It is very likely that the calculated values of intensity given by Spear and Grimmer<sup>(1)</sup> are more nearly correct than those obtained by measurement.

#### § 5. ACKNOWLEDGEMENTS

The author wishes to acknowledge his indebtedness to Mr N. H. Pierce for his assistance with the experiments, and to the British Empire Cancer Campaign whose financial support has helped make it possible to carry out the work.

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#### DISCUSSION

Mr W. BINKS. It is now well known that in order to measure gamma radiation by an ionization method, the walls of the measuring-chamber should be thick enough to prevent extraneous primary and secondary beta radiation from entering the air cavity—that is to say, the thickness of the chamber-wall should not be less than the range of the fastest beta particles in the material of which the wall is composed. In this connexion I would like to draw the author's attention to a number of papers published in recent years by Friedrich, Schulze and Zimmer, Glasser, Mayneord and Roberts and by Kaye and myself. In the case of chambers having walls thinner than the beta-ray range the ionometric conditions are somewhat complex, the measured ionization depending upon the particular experimental arrangement adopted. For example, it seems to depend not only on the material and thickness of the filter but on the latter's position—that is, on whether the filter is near the radium or near the chamber. Further, it depends on the size and shape of the source and chamber and on the distance between them.

The observations of Friedrich and Schulze (1935) on the effects of different filters (0.1 mm. platinum, 0.3 mm. platinum plus 1.5 mm. brass, and 6.6 mm. lead) would appear to contradict those of Mr Wilson, but it should be noted that the

sources and chambers and their distances<sup>1</sup> apart were different in the two cases. Thus the ionization measured by means of chambers having too thin walls is dependent upon the distribution of the primary and secondary beta radiation at the location of the chamber, the curves shown by Mr Wilson representing a particular example of this phenomenon.

I agree with the author that it would be impracticable to make reliable measurements of gamma radiation at distances from the source less than the range of the beta particles. But it would seem that when considering the biological effect of the radiations from radium one ought to take into account the beta radiation as well as the gamma radiation received by the tissue. In certain locations, for instance in the case of tissue adjacent to a radium needle or that of the superficial layers of tissue of a patient who is irradiated from an external radium source, the secondary beta radiation has an important bearing on the total energy absorbed by the tissue. It is doubtful, therefore, whether the calculated gamma-ray intensities at distances less than 4 mm. from a radium source give even an approximate idea of the actual energy absorbed. For these distances it would be better to use ionization chambers of appropriate wall-thickness.

AUTHOR'S reply. I was quite aware of the fact that other workers had dealt with the problem of secondary beta radiation, but it was just this particular example of the phenomenon with which I was concerned. The work done by Spear and Grimmett was carried out in this laboratory, and since it dealt with an important biological effect that occurred for a specific gamma-ray intensity, it was essential to know the value of the latter. The experiments I have described seem to show which of the intensity-values published by Spear and Grimmett was correct, and for this reason they appeared to be worth recording.



# ON THE OPTICAL CONSTANTS OF ALLOYS OF THE COPPER-ZINC SYSTEM

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*Received 13 February 1937. Read 9 April 1937*

**ABSTRACT.** Values of the optical constants (refractive index, reflection coefficient and absorption coefficient) are given for some alloys of the copper-zinc system ( $\alpha$ ,  $\beta$  and  $\gamma$  brass) over the range  $\lambda$  4358 to  $\lambda$  6800, as determined by the method of Drude. Curves showing the variation in the optical constants with composition for particular wave-lengths of light are also shown.

## § 1. INTRODUCTION

IN continuation of an experimental investigation on the optical constants of some simple binary alloys<sup>(1)</sup>, the present paper is concerned with dispersion-measurements in the visible region of the spectrum for the copper-zinc system.

This system is interesting from an experimental point of view on account of the extension of the absorption band of copper into the visible, resulting in the very distinct colour-variation in alloys in the  $\alpha$  phase. From the red of pure copper the colour changes gradually to the typical yellow of brass at the end of the  $\alpha$  phase (39 per cent by weight of zinc). The narrow  $\beta$  phase (46.5 to 49.6 per cent of zinc) again shows the red of copper, and from 60 to 69 per cent of zinc we have the extremely hard, brittle, silvery  $\gamma$  phase.

Measurements of the optical constants of alloys in each of these three phases have been made, though the main part of the work has been concerned with the  $\alpha$  phase. An attempt has been made to obtain dispersion curves over the range  $\lambda$  4358 to  $\lambda$  6800 for a number of specimens of varying composition, and to obtain curves showing the variation in the optical constants with composition for particular wave-lengths of light.

## § 2. EXPERIMENTAL DETAILS

The general arrangements of the apparatus and method used, essentially that of Drude, have been fully described in previous papers. A parallel beam of light, plane-polarized at an azimuth of  $45^\circ$  to the vertical, is incident at an angle  $\theta$  on a polished specimen of the alloy mounted vertically. The reflected elliptically polarized light is analysed by a Soleil-Babinet compensator, measuring the phase-difference  $\Delta$  between the horizontal and vertical components of the beam, and an

analysing Nicol prism fitted with a half shade to measure the azimuth  $\psi$  of the restored plane-polarized light.

From the measured quantities  $\theta$ ,  $\Delta$  and  $\psi$  the refractive index  $n$  and the absorption coefficient  $K$  are calculated from the following equations, due to Drude:

$$\begin{aligned}\tan Q &= \sin \Delta \tan 2\psi, \\ \cos 2P &= \cos \Delta \sin 2\psi, \\ S &= \sin \theta \tan \theta \tan P, \\ A &= S^2 \cos 2Q + \sin^2 \theta = n^2 - K^2, \\ B &= S^2 \sin 2Q = n^2 + K^2, \\ 2n^2 &= \sqrt{(A^2 + B^2)} + A, \\ 2K^2 &= \sqrt{(A^2 + B^2)} - A.*\end{aligned}$$

The reflecting-power at normal incidence is given by

$$R = \frac{n^2 + K^2 + 1 - 2n}{n^2 + K^2 + 1 + 2n} \times 100 \text{ per cent.}$$

In view of the decay in the measured phase-difference  $\Delta$  with exposure, corrections have been applied to this constant throughout the work in this paper. The corrections were applied to the compensator scale readings from observations taken at intervals during the experiments. All values of  $\Delta$  given in the tables were calculated from these corrected compensator readings.

*Preparation of the specimens.* The copper specimens used were prepared from clean electrolytic copper, melted and allowed to solidify in vacuo. The brasses were prepared in air in an induction furnace. Electrolytic copper was first melted and the zinc, comprising B.D.H. zinc sticks As.T., was dropped into the melt in small pieces, each wrapped in paper to retard volatilization and ignition until the zinc was submerged. By using an excess of zinc to compensate for the loss in melting, alloys of approximately the desired compositions were obtained. The alloys cast cleanly into iron moulds and were quite free from pinholes.

Chemical analysis of the specimens gave the percentages by weight of copper shown in the table.

Composition of specimens

Specimen number Phase	1 $\alpha$	2 $\alpha$	3 $\alpha$	4 $\alpha$	5 $\alpha$	6 $\beta$	7 $\gamma$	Extra specimen, $\alpha$ phase
Weight of copper (per cent)	93.42	91.09	87.60	80.60	74.61	52.85	32.75	86.12
Weight of zinc (per cent) by difference	6.58	8.91	12.40	19.40	25.39	47.15	67.25	13.88
Atomic percentage of zinc	6.42	8.67	11.92	18.98	24.85	46.45	66.63	13.56

\* The imaginary dielectric constant  $\epsilon$  occurring in dispersion theory of metals is written  $\epsilon = (n - iK)^2$ , where  $n$  and  $K$  are as above. Drude used  $\epsilon = n^2(1 - ik)^2$  so that his absorption coefficient  $k = K/n$ .



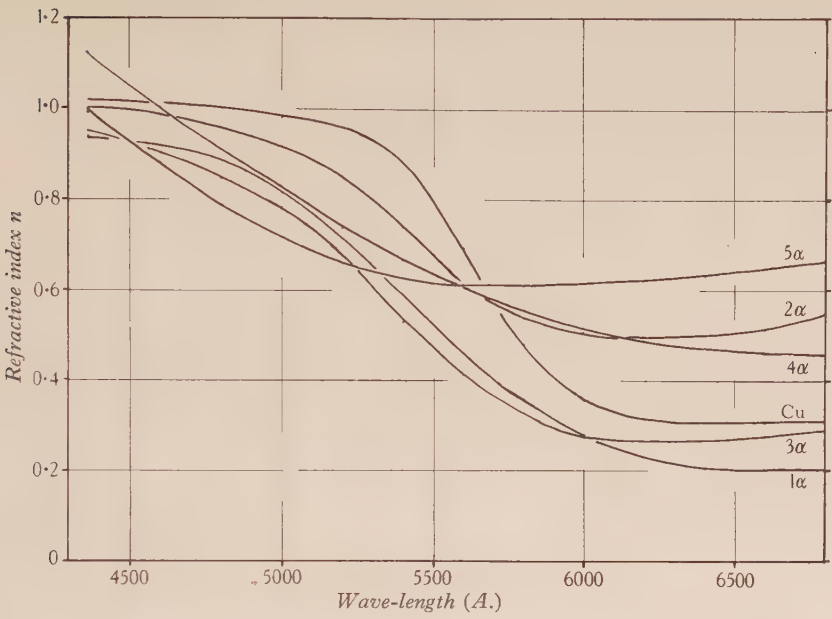


Figure 1.

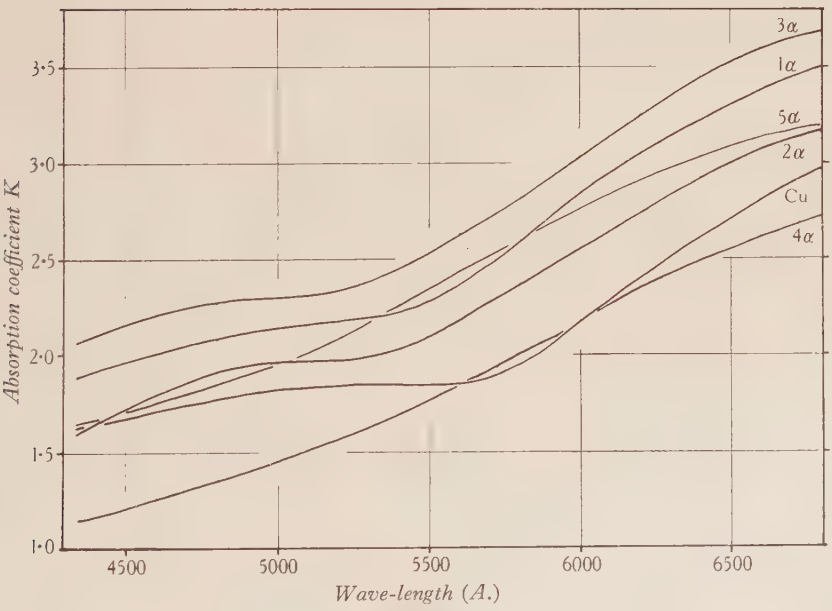


Figure 2.

Micrographic examination of the specimens revealed a cored structure in the  $\alpha$  specimens. This was removed by annealing at  $580^{\circ}$  C. for 40 hours.

The mirror surfaces were prepared, after a preliminary grinding with a 0000 grade of emery paper, by polishing on Selvyt cloth with magnesia. The values of the optical constants for each specimen for a range of wave-lengths in the visible spectrum are plotted against wave-length in figures 1 to 4.

### § 3. DISCUSSION

Though the variation of the constants with wave-length for individual specimens yields smooth curves in every instance, there is no correlation between dispersion curves for specimens of different composition. Attempts to select, for given wave-lengths, series of values of the constants from the dispersion curves, figures 1 and 2, and to plot these against composition result in curves entirely devoid of any distinct form. This lack of coherence is a consequence of differences in the polished surfaces; results of this kind illustrate the difficulties attendant upon the study of the optical properties of an alloy system. If the constants determined for each specimen were truly characteristic of the metal or alloy and were not influenced by the state of the surface, a natural coherence would occur between the results for different specimens. Alternatively, a similar coherence would be obtained if a process of polishing could be devised to produce the same surface condition in each alloy, though the constants would not necessarily represent the optical properties of the undisturbed metallic structure; see figures 5 and 6.

In the case of the copper-nickel alloys<sup>(1)</sup> some success was attained in producing the same polish with each alloy and, by averaging the results from three separate polishes in order to even out minor irregularities, smooth {constants, composition} curves were obtained. The difficulty as regards repeating this successful procedure in the present instance lies in the fact that whereas the copper-nickel alloys are hard and relatively easy to polish, the brasses are soft and easily scratched; moreover, their colour makes it more difficult to discern a slight tarnish which sometimes appears on the surface during polishing. To polish a piece of copper or soft brass to a clear glass-like state of surface by the usual methods, avoiding excessive cold-working of the surface, is an almost impossible task. Very fine scratches nearly always appear<sup>(2)</sup> and, as Drude has shown, these are capable of polarizing light, particularly when they are unidirectional. Though the measured phase-difference  $\Delta$  between the components of the reflected elliptically-polarized light is not influenced by these scratches, the azimuth  $\psi$  is increased for scratches parallel to the plane of incidence and decreased for scratches perpendicular to the plane of incidence.

This variation can be observed experimentally. It is of the order of  $1^{\circ}$ . Discrepancies due to it may be avoided by carrying out the final stages of polishing so as to avoid unidirectional scratches or by taking care always to mount the specimen with the predominating scratch-directions always the same.



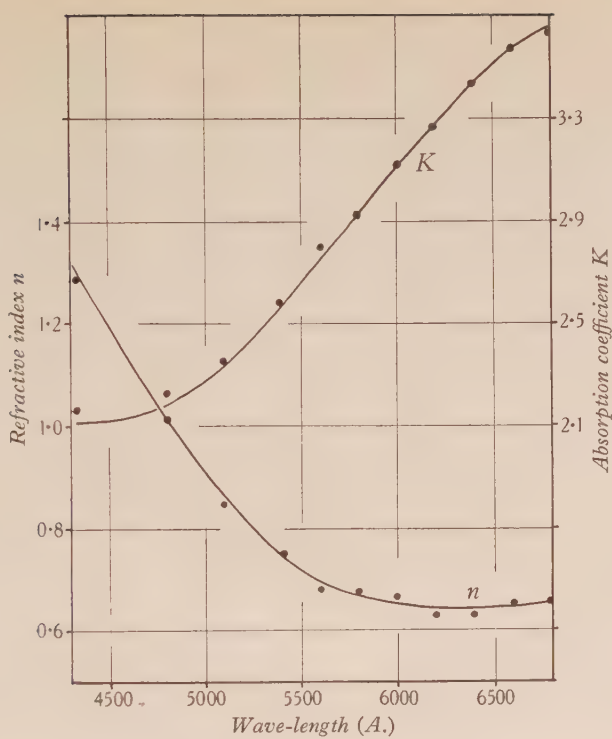


Figure 3. Specimen  $\beta$  brass.

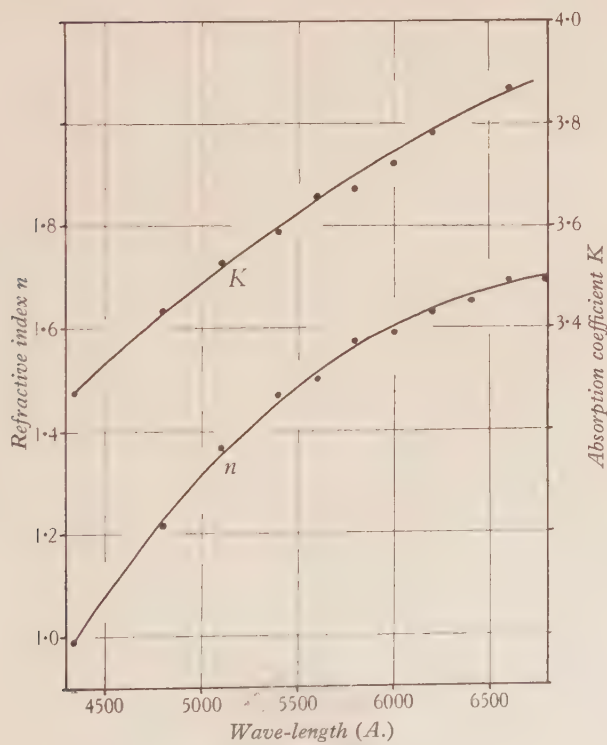


Figure 4. Specimen  $\gamma$  brass.

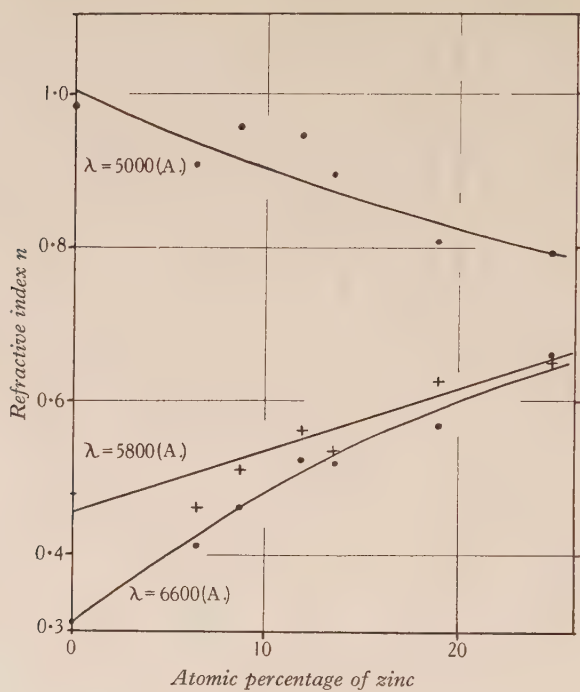


Figure 5.

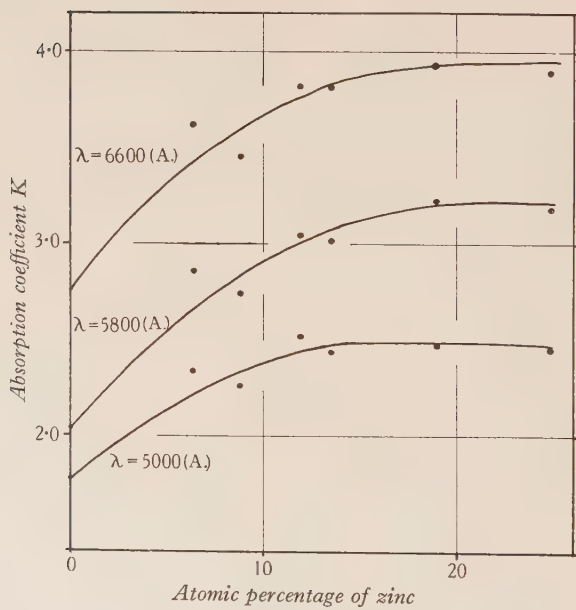


Figure 6.

The possibilities of using the electrolytic polishing process of Jacquet<sup>(3)</sup>, which has proved successful for copper<sup>(4)</sup>, were considered. Here again the position is not satisfactory. Though the method is quite feasible for brasses, it is more tedious in operation since the exact conditions of solution and current and voltage have to be determined separately for each alloy. Again, there is no reason to believe that these conditions, when correctly determined and applied, would effect the removal of the constituents of the alloy (made the anode in the process) in the proportions in which they happened to be present. Finally, the considerable variation found in the

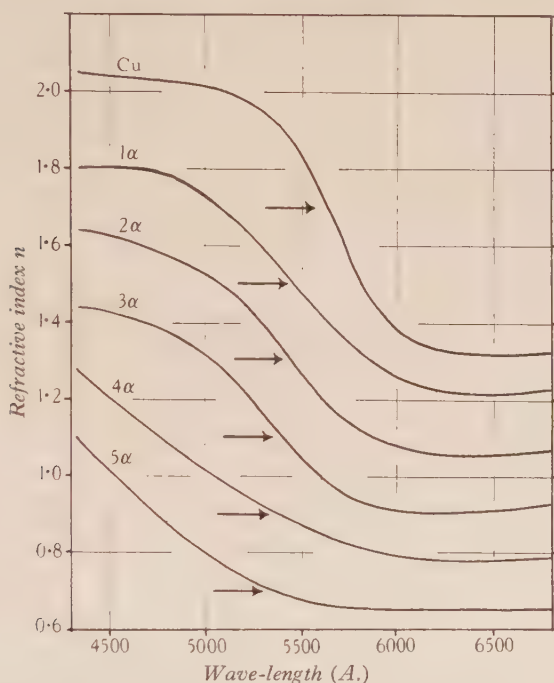


Figure 7. To obtain refractive indices from the graph the following corrections should be subtracted from the ordinates: 1.0 for copper, 0.8 for 1α, 0.6 for 2α, 0.4 for 3α, 0.2 for 4α, 0.0 for 5α. The arrow indicates the value 0.7 of  $n$ .

measured constants when different depths of the mechanically smooth disturbed surface are removed introduces the disadvantage of having to make an arbitrary choice of the depth of material to be removed, and this in turn entails much laborious work to ensure that the depth shall be the same in every instance.

The method employed in the present work is that of embedding all the specimens in a lead block and grinding and polishing them simultaneously. This ensures that each one receives practically the same treatment as regards the amount of cold work sustained. Even so, however, it is hardly probable that alloys with slightly different mechanical properties will receive identical treatment, and the optical constants measured on specimens polished in this way do not give as smooth a curve when plotted against the zinc-content as might be expected for a variation of a



physical constant produced by an increasing proportion of zinc in a solid solution. The variation of  $n$  and  $K$  with composition is shown in figures 5 and 6 for three wave-lengths,  $\lambda$  6600,  $\lambda$  5800 and  $\lambda$  5000 Å., for the alloys 1 $\alpha$ , 2 $\alpha$ , 3 $\alpha$ , 4 $\alpha$ , 5 $\alpha$ , simultaneously polished in the lead block.

It is evident that the dispersion curves, figures 1 and 2, though showing an increasing divergence in form from the curves characteristic of copper as the zinc-content increases, require correction of the constants before any quantitative

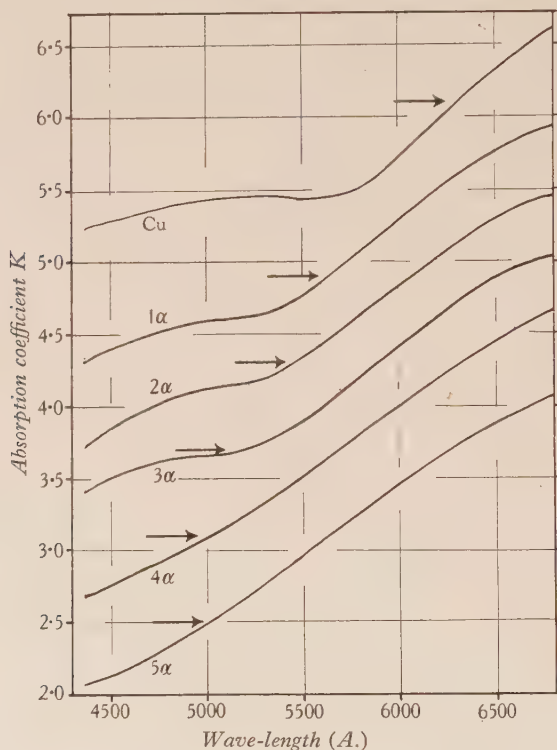


Figure 8. To obtain refractive indices from the graph the following corrections should be subtracted from the ordinates: 3.6 for copper, 2.4 for 1 $\alpha$ , 1.8 for 2 $\alpha$ , 1.2 for 3 $\alpha$ , 0.6 for 4 $\alpha$ , 0.0 for 5 $\alpha$ . The arrow indicates the value 2.5 of  $K$ .

relationship will appear between them. It was, in fact, principally with the object of suggesting quantitative corrections which could be applied to these dispersion curves in order to locate them in a reasonable relationship that the change in the constants with composition was investigated.

Variations in optical constants due to differences in polish take the form of an approximately parallel shift of the dispersion curves to higher or lower values of the constants. Figures 7 and 8 show the dispersion curves for  $n$  and  $K$  after the curves for each alloy have been so shifted to such an extent that the values of the constants for the wave-lengths  $\lambda$  6600,  $\lambda$  5800 and  $\lambda$  5000 Å. now coincide with the

values read off from the smooth curves drawn in figures 5 and 6 for the variation of the constants with composition.

It must be clearly understood that this arrangement of the results is not intended to do more than indicate the interrelationship between the constants of copper and the  $\alpha$  brasses shown by our experiments. The values of the constants are correct values only in so far as the constants for the mechanically polished copper specimen may be assumed to be true values. That these may vary considerably upon repetition and with different polishing technique is well established and has been discussed more fully in a previous paper<sup>(4)</sup>.

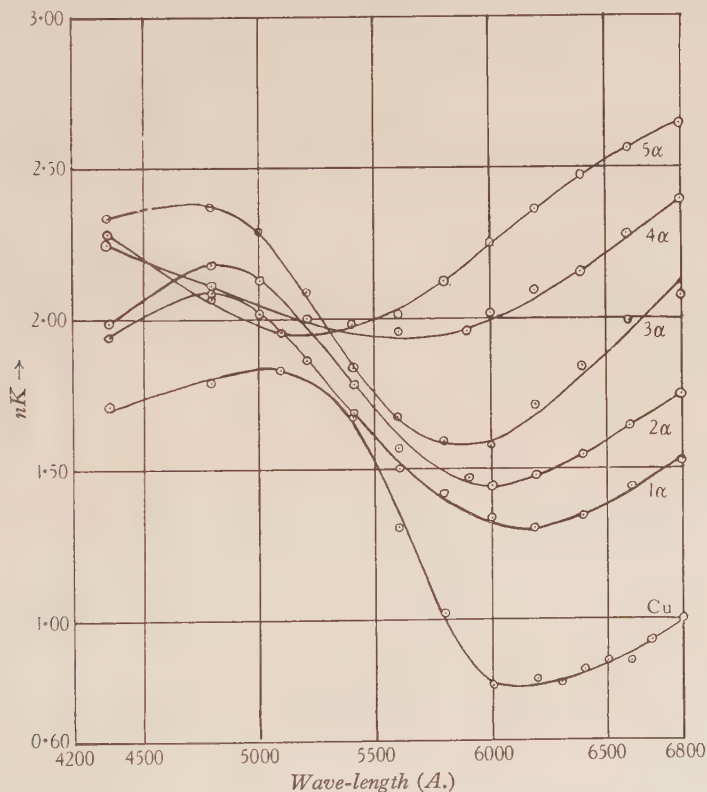


Figure 9.

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# THE THEORETICAL INTERPRETATION OF THE OPTICAL CONSTANTS OF COPPER- ZINC ALLOYS

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**ABSTRACT.** The results of the experiments of Lowery, Wilkinson and Smare on the optical constants of alloys of copper and zinc ( $\alpha$ ,  $\beta$  and  $\gamma$  brass) are discussed from the point of view of the electric theory of metals.

**I**N this note we discuss the results of the preceding paper<sup>(1)</sup> from the point of view of the electric theory of metals.

If  $n$ ,  $K$  are the optical constants, then  $nK$  is related to the rate of absorption of energy by the metal in the following way. If we consider a volume of metal  $d\tau$  in which the electric vector of the light is  $E$ , then<sup>(2)</sup>

$$nK\nu d\tau = \frac{\text{rate of loss of energy}}{E^2}.$$

In figure 9 of the preceding paper are shown the values of  $nK$  for the copper-zinc alloys. Pure copper is seen to have an absorption band with its long wave-length limit between  $\lambda$  5500 and  $\lambda$  6000. It has been suggested<sup>(2)</sup> that this absorption band is due to the ejection of electrons by the incident light from the  $3d$  level of the copper atom. The type of electronic transition responsible is thus exactly the same as that which gives rise to X-ray absorption edges; as we know from the chemical behaviour of copper, the energy required to remove an electron from the  $d$  level is very small, so it is not surprising that the absorption edge lies in the visible. The fact that the absorption edge is not sharp may be ascribed to a broadening of the  $3d$  level of the atom into a narrow band of levels in the crystal.

As zinc is added to copper, three effects may be noticed, as in figure 9 of the previous paper. (a) The absorption on the long wave-length side of the edge increases. (b) The absorption edge becomes less sharp. (c) The absorption edge is displaced in the direction of shorter wave-lengths.

Effect (a) can be ascribed simply to the increased resistance of the metal, which damps the induced vibrations of the electrons in the metal and so increases the absorption. We shall not attempt to calculate the absorption to be expected.

Effect (b) can be accounted for as follows. In order to find the energy  $h\nu$  corresponding to the wave-length of the edge we have to calculate the work re-



quired to move an electron from the  $3d$  level of one of the copper atoms to the conduction levels of the crystal. This is made up of two parts: (1) The work  $W_1$  required to remove an electron from a  $d$  level of a copper atom right out of the crystal. (2) The work  $W_2$  (*minus* the work function of the alloy) required to bring the electron back into the crystal. We consider first the term  $W_1$ . The picture of a copper-zinc alloy to which one is lead by the electron theory of metals is an array of ions  $\text{Cu}^+$  and  $\text{Zn}^{++}$  embedded in a sea of negative charge. The negative charge tends to cluster round the zinc ions so that each copper atom has only a small excess negative charge and each zinc atom a small excess positive charge. These charges have been estimated from the resistivity<sup>(2)</sup> and from the energy of the super-lattice<sup>(3)</sup> to be about  $\Delta Q = \pm 0.075e$ , where  $e$  is the electronic charge. Now the electrostatic potential at any lattice point due to these charges will on the average be zero, because the copper and zinc atoms are distributed at random in the lattice. But the electrostatic potential at any copper atom which happens to have more zinc atoms as nearest neighbours than the average number will be higher than zero, and at those which have an excess of copper atoms as nearest neighbours it will be lower. The potential of any lattice point will fluctuate by an amount comparable with  $\Delta Q/r_0$ ,  $r_0$  being the interatomic distance. Thus the work required to remove an electron from the lattice will fluctuate by an amount of the order of magnitude  $e\Delta Q/r_0$ , which is approximately equal to  $0.075e^2/r_0$  or  $0.4$  eV. Since a wave-length  $\lambda$  5500 corresponds to 2.2 eV. this is of the order of magnitude which is observed.

Effect (c). According to the electron theory of metals the kinetic energies of the electrons in a metal have values between 0 and a maximum energy  $E_{\text{max}}$  given by

$$E_{\text{max}} = 36.1 (n_0/\Omega)^{\frac{2}{3}},$$

where  $\Omega$  is the volume per atom in  $\text{cm}^3 \times 10^{24}$  and  $n_0$  the mean number of free electrons per atom. If an electron is added to the metal, it cannot have kinetic energy less than  $E_{\text{max}}$ . Thus we may write for the work  $W_2$  required to bring an electron back into the metal

$$W_2 = \text{a negative constant} + E_{\text{max}}.$$

Now in a series of copper-zinc alloys, as the amount of zinc is increased  $n_0$  increases much faster than  $\Omega$  and so  $E_{\text{max}}$  increases. The values are as shown in table 1.

Table 1

Composition of alloy (Atoms per cent of zinc)	$n_0$	$E_{\text{max}}$ (eV.)
0	1	6.98
20	1.2	7.70
40	1.4	8.33
50	1.5	8.61

Thus for an alloy containing 20 atoms per cent of zinc, the absorption edge will be shifted towards shorter wave-lengths by

$$7.70 - 6.98 = 0.28 \text{ eV.}$$

Taking the wave-length of the edge for pure copper to be 6000 Å., the theoretical values for the alloys shown in figure 9 of the preceding paper are as shown in table 2.

Table 2

Alloy	Composition (Atoms of zinc per cent)	Shift in the edge (eV.)	Absorption edge (calculated)
1α	6.42	0.2	λ 5500
2α	8.67	0.3	5250
3α	11.92	0.4	5000
4α	18.98	0.6	4640
5α	24.85	0.8	4300

The displacements of the edge are rather greater than those observed; the latter are however difficult to estimate, owing to the broadening of the edge discussed above.

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# SENSITIVITY VARIATION OF X-RAY PHOTOGRAPHIC FILMS

BY WM. H. GEORGE, M.Sc., Ph.D., F.Inst.P., University of Sheffield

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**ABSTRACT.** An experimental study of the variation of sensitivity from place to place on an X-ray photographic film has been made, so that fluctuations of blackening, due to intensity-variation of the incident radiation, can be distinguished from those due to film sensitivity. Double-film exposures are made with the two sensitive surfaces in contact, and the processing is carried out in a simple mechanical apparatus so that the films move always with accelerated motion relative to the liquids. It is found that if X-ray photographic films are used with the technique described and are blackened until they transmit about half the incident light, then the variations of sensitivity are such as may lead to maximum deviations in the individual measurements of X-ray intensity of the order  $\pm 6$  per cent in single-film exposures or of  $\pm 3$  per cent in double-film exposures. The mean deviations are of the order  $\pm 2$  per cent.

## § 1. INTRODUCTION

THE results of the experiments here described relate to the following problem. If a piece of X-ray film exposed in a uniform beam of X rays is then processed (i.e. treated) under prescribed conditions, to what degree of accuracy will adjacent regions of the film be uniformly blackened?

Although the problem arose out of some work on the interaction of polarized X rays and crystals<sup>(1)</sup>, the results are applicable also to X-ray crystallography with tiny single crystals giving small spots in rotation and oscillation photographs, or to fine spectral lines in powder photographs.

## § 2. EXPERIMENTS

*Uniform exposure.* The object of the work is, in the measurement of a fluctuating X-ray intensity, to differentiate between the X-ray fluctuations and those due to the photographic materials and processes. Since an X-ray beam cannot be directly observed by the coincidence method, the uniformity of the beam cannot be directly judged. In any very sensitive laboratory method of measuring X-ray intensity, the final observations, consisting of deflections of some measuring instrument, may fluctuate about a mean value but are never constant when the ionization chamber or other detecting apparatus is moved across the beam of X rays. It is therefore necessary to define in terms of observable quantities what part of the fluctuations of blackening of a film are to be ascribed to variations of intensity of the incident beam.



Two facts help in formulating the definition. Firstly, it is found that the time average of intensity at some distance from an X-ray tube is free from rapid fluctuations in adjacent tiny areas (say  $1 \times 0.2$  mm.). Secondly, the absorption of X rays in photographic film is so slight that the beam transmitted through one film is still sufficiently intense to blacken a second film placed behind and in contact with the first film. Hence if two films are exposed in contact and at some distance from an X-ray tube, any difference between the ratio of blackening of two areas on the first film and the corresponding ratio on the second film will be ascribed to variation in the film-sensitivity. The mean blackening for the two areas will be fixed by using the first fact in the way explained in § 3.

All the experiments were therefore carried out by exposing simultaneously pairs of films in contact. The camera is shown in figure 1. The two films, protected from light by black paper, rest behind two lead screens pierced at front and back by a slit *S* exposing a suitable area of film and allowing the X-ray beam to pass through this part of the camera and its support at the back. The time *t* of exposure necessary to blacken a film until it transmitted only half of the light incident normally upon it in a microphotometer was first determined. The camera was then loaded with a pair of experimental films and set upon its geometrical supports *A*, *B*, *C*. When the X-ray tube was running steadily, two exposures were made, each for a period of a little more than  $\frac{1}{2}t$ . During the first period the camera was fixed in the position shown in figure 1, and during the second it was reversed about a vertical axis through *A*. The slit therefore remained in the same position relative to the X-ray tube but during the second period the beam passed through the films in the reverse direction. The exposure for each film was therefore  $\frac{1}{2}t$  in the direct beam and  $\frac{1}{2}t$  in a beam which had first penetrated the other film. The same region of the beam was used in each period for the same part of each film.

For intensity-calibration the films were then removed from the camera and a rotating-sector exposure was made in another camera upon a part of the unexposed surface.

A filament X-ray tube with copper anticathode and nickel filter was used in all the experiments.

*Uniform development.* Uniform development as usually defined seems to be inapplicable to experimental work when different parts of a film are exposed in a uniform beam for different times, or when they are exposed in a non-uniform beam for the same time or in fact in any circumstances resulting in non-uniform blackening. Suppose that developer kept at constant temperature and concentration is

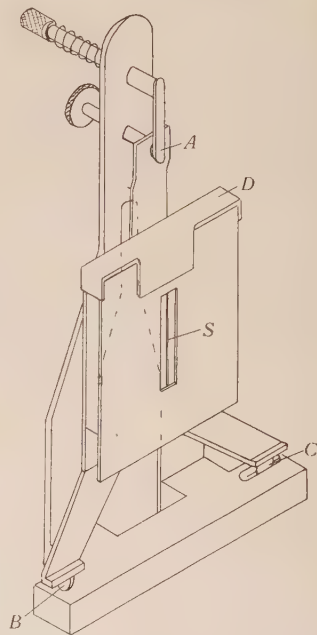


Figure 1. Diagram of camera. *S*, slits; *A*, *B*, *C*, geometrical constraints; *D*, lead cap.

directed on to such a film in a rapid uniform stream. The developing chemicals will diffuse into the gelatin and react with the affected silver-halide grains, producing various reaction products which diffuse outwards into the stream of fresh developer. Within the film the rate of blackening will depend upon the concentration of developing chemicals, and upon the diffusion rates of their replacement and of the removal of reaction products. These constants are beyond the experimenter's control. In much optical photography, including sound-recording on film, only a thin layer on the upper surface of the emulsion is affected by the radiation, but in the present experiments the absorption of the X rays is so slight that the whole depth of the exposed regions contains developable grains. The chemicals must therefore diffuse through thicker regions than in optical photography. According to common supposition the diffusion must take place also through a layer of liquid adhering to the surface of film in contact with the moving liquid.

Because difficulties arising from the grain and associated with extra background fog, a film which is to be measured on a microphotometer is not developed to gamma infinity. If, therefore, adjacent patches of the film have first been subject to different exposures, it follows that during development they will not be in contact, *within* the film, with the same concentration of developing chemicals for the same time. In this sense then uniform development is impracticable. Since experimental research is concerned with the accessible rather than with the inaccessible it is more helpful here to choose some other definition of uniform development.

Instead of the arbitrarily chosen theoretical definition of uniform development an arbitrarily chosen definition based on the experimenter's actions<sup>(2)</sup> will be used. Accordingly a film will be said to be subject to uniform development when, at constant temperature, its whole surface has been immersed throughout in rapidly flowing developer of constant composition or in violently stirred developer whose composition is changed only by the progress of the development reactions. A method by which these conditions may be fulfilled for a fairly large film, coated with emulsion on one side and blackened optically, is described elsewhere<sup>(3)</sup>, together with a survey of many other methods. There is considerable difficulty in using the method for X-ray films measuring only  $3 \times 2$  cm. and coated on both sides with emulsion. After trial of a number of different methods the simple apparatus shown in figure 2 was devised; it was found to give self-consistent results.

*Processing.* The two films are firmly clamped side by side vertically in a detachable framework *F* made of ebonite and stainless steel. The glass tank *T* and the block *B* are removable, but the rest of the apparatus, consisting of reciprocating mechanism, reduction pulleys and electric motor, is screwed to the bench. To avoid delay in making the necessary changes, a separate tank is used for each liquid. When the loaded frame is fixed in position the dry films are quickly plunged into developer by raising the tank, and the reciprocating mechanism is started. This gives 140 complete vibrations per minute, each of total displacement 2 cm.

The films always move with accelerated motion relative to the solutions. This was relied upon to help the interchange of liquids at the film surface. To change the surrounding liquid, a propeller from a toy motor boat was rotated in front of the

oscillating films. As the results obtained with and without the use of the propeller appeared to be the same, it was later discarded. The agitation of the liquid was studied by two methods. The first was with coffee grounds, washed and decanted several times to remove all turbidity. These showed the large-scale agitation to be very thorough and free from dead regions. Secondly the regions near the film were examined in close detail by using dye introduced through a fine jet. The removal of liquid introduced as close to the film as possible and the dispersal throughout the bulk of liquid were too rapid to be followed by eye. In all the experiments splashing and the introduction of air bubbles, which accelerate oxidization of the developer, were avoided by adjusting the depth of liquid in the tank.

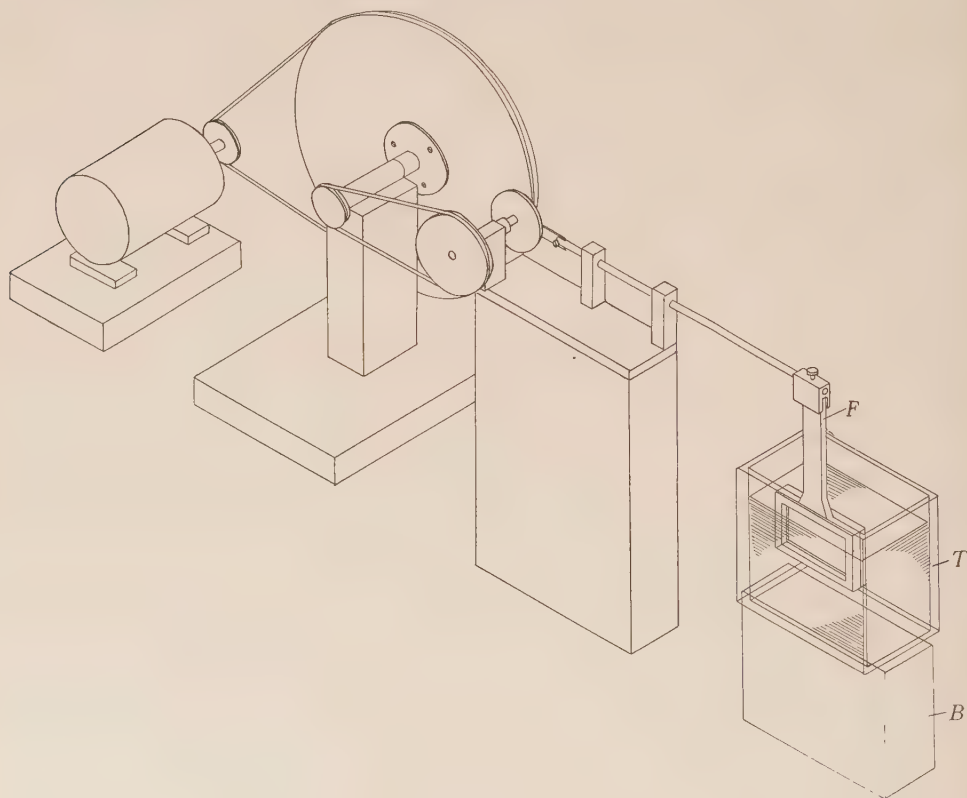


Figure 2.

The routine followed was to develop the film for 5 min. at 18° C. in a solution containing, per litre: Elon 2 g., sodium sulphite (desiccated) 72 g., hydroquinone 8.8 g., sodium carbonate (desiccated) 48 g., potassium bromide 4 g. After being washed for 3 minutes, the film was fixed for 15 minutes in a solution containing, per litre: hypo 240 g., sodium sulphite (desiccated) 10 g., sodium bisulphite 25 g.

The films were then washed for 3 minutes and hardened for 3 minutes in a solution containing, per litre, 10 cm. of formalin (40 per cent formaldehyde solution) and 5 g. of desiccated sodium carbonate. Next, the frame was removed from the



oscillator and the films were washed in running water for from  $1\frac{1}{2}$  to 2 hours. From time to time the frame was agitated in the water to remove the bubbles which collect on the film surfaces even when rapidly flowing water is used. When the washing was completed, viscose sponge was used to remove as much water as possible from the films which were then dried slowly in a horizontal position at room temperature. Filtered solutions were used throughout to avoid scratching of the film surfaces by particles thrown against them by the rapidly moving liquids.

*Microphotometry.* The dry films were measured with a Moll microphotometer<sup>(4,5)</sup>. To ensure that the galvanometer deflections should be, as far as possible, unaffected by deviation of light out of the optic plane containing the thermopile slit, owing to scratches, the viscose sponging of the film was always done in a direction perpendicular to that of its subsequent passage through the microphotometer.

Each instantaneous galvanometer deflection was the result of exploring a region of the film measuring  $1 \times 0.033$  mm.

### § 3. RESULTS

On account of the graininess of the film, the record of galvanometer deflections taken on the microphotometer drum *D* appears as a line subject to many sudden inflections. Each record was therefore divided into strips 5 mm. wide, from which the readings plotted in the two typical pairs of graphs, figure 3, were obtained as follows. The films were exposed in pairs and each half of the figure represents the

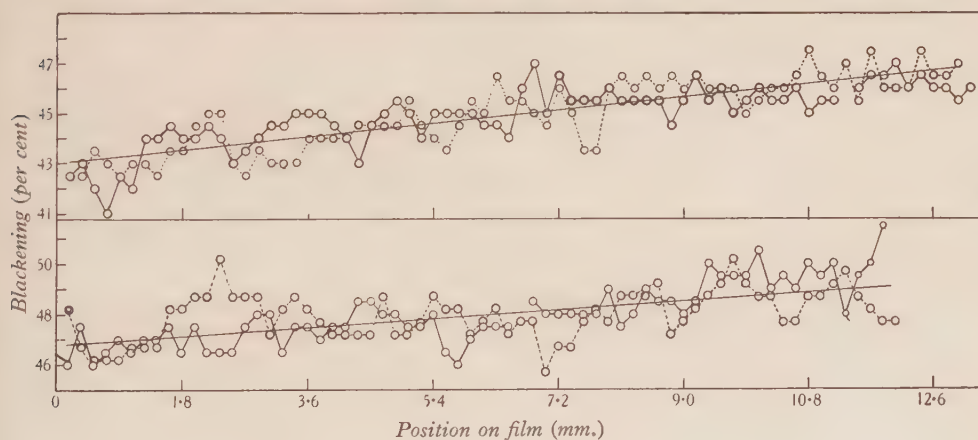


Figure 3.

results got from one pair. Points got from measurements of one film of the pair are joined by solid lines. Broken lines are used to join the points relating to the other film of the pair. A straight line was drawn through each portion of the whole curve contained in each 5-mm. strip of the paper record got from the microphotometer drum, so as to divide its curve into two parts lying equally above and below the straight line. The galvanometer deflection corresponding to the mid-point of this

line was taken as the average of the whole 5-mm. strip. To avoid possible error due to prejudice, strips adjacent to the one studied were hidden from view.

The measurements were expressed in terms of the blackening  $B$  defined as follows:

$$B = \frac{\text{galvanometer deflection due to strip of blackened film}}{\text{galvanometer deflection due to clear film}} \times 100;$$

with this definition the blackening of an opaque film would be 100 per cent, of a clear film zero, and of a film transmitting one-half of the light incident normally upon it 50 per cent. Each point on the graphs corresponds to a strip 5 mm. wide on the drum record  $D$  or to a region measuring  $1 \times 0.18$  mm. on the X-ray film. Since the double exposure of each pair of films was made without disturbing the films in their holder, the regions of each in contact could be determined by the distance from a shadow cast by a lead screen behind which part of each film was protected from the X rays. Each unit of displacement on the horizontal axis of figure 3 represents a displacement of 0.18 mm. on the film.

Each of the straight lines, later taken as representing the blackening due to the X-ray intensity to be measured, was got from a mean curve passing through the algebraic mean of each pair of points, one from the solid and one from the dotted curve. For the upper diagram the seventy points of this mean curve were then divided into sets of ten adjacent points and the algebraic mean of each set was used to give one point. The method of least squares was used to draw the straight line through the six points. In the lower diagram the points were grouped into seven sets of eleven each.

An analysis of the deviations of the observations from this derived mean due to sensitivity variation of X-ray film is shown in table 1.

Table 1. Numbers of points representing deviation from mean blackening

	Limits of variation (per cent)					
	$\pm(0 \text{ to } 0.5)$	$\pm(0.5 \text{ to } 1.0)$	$\pm(1.0 \text{ to } 1.5)$	$\pm(1.5 \text{ to } 2.0)$	$\pm(2.0 \text{ to } 2.5)$	$\pm(2.5 \text{ to } 3.0)$
Upper diagram:						
Dotted curve (i)	39	22	6	1	1	—
Solid curve (ii)	36	25	6	3	—	—
Mean of paired points from (i) and (ii)	47	20	2	—	—	—
Lower diagram:						
Dotted curve (i)	35	21	6	2	1	—
Solid curve (ii)	31	17	10	6	1	1
Mean of paired points from (i) and (ii)	41	20	4	—	—	—

The use of simpler methods of processing the films, such as developing in a rocking dish, or stroking with cotton-wool under the liquids or rotating in a flask of liquid, gave fluctuations much greater than those shown in table 1, and the results were too inconsistent to justify their being reported. The results given in figure 3 are typical provided the technique described in this paper is used. When it was

found that the method gave self-consistent results, further experiments were discontinued. To establish which parts of the technique may be omitted without affecting the results an elaborate series of control experiments of the cause-and-effect type would be necessary.

The following conditions apply to all the results reported. The exposures were made with general radiation from an X-ray tube with a copper anticathode and a nickel filter. The photographic density of the exposures was about 0.301; the area of blackening was about  $1 \times 12$  mm.; the individual mean measurements relate to tiny strips  $1 \times 0.18$  mm. From rotating sector photographs it was found that under these conditions a variation of blackening of 1 per cent corresponds to a change of X-ray intensity of 2 per cent. The variations of sensitivity are summarized in table 2.

Table 2. Percentage deviation from mean blackening (50 per cent) or density (0.3)

	Single-film exposure	Double-film exposure
Maximum departure by single measurements from mean blackening (per cent)	Blackening 3 X-ray intensity 6	Blackening 1.5 X-ray intensity 3
Percentage of measurements showing this maximum departure	1	4
Percentage of measurements lying between 49.5 per cent and 50.5 per cent	50	65
Percentage of measurements lying between 50.5 per cent and 51 per cent or between 49 per cent and 49.5 per cent	30	30

From this summary it is evident that with the use of the technique described, the variations of sensitivity of X-ray film are such as may lead to maximum deviations in the individual measurements of X-ray intensities of the order  $\pm 6$  per cent in single-film exposures or of  $\pm 3$  per cent in double-film exposures. The mean deviations are of the order  $\pm 2$  per cent.

#### § 4. ACKNOWLEDGEMENTS

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# RAPID VARIATIONS IN THE MAGNETIC FIELD PRODUCED BY LIGHTNING DISCHARGES

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Communicated from the Institute of High Tension Research of  
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**ABSTRACT.** Measurements of the rapid variations in the magnetic field produced by lightning discharges have been made by means of a frame aerial and a cathode-ray oscillograph. The observations made it possible under certain conditions to calculate the current-variations in lightning discharges, and maximal values up to 100 kA. were derived. These were in good agreement with results obtained by other indirect methods. Conclusions as to the structure of the current-variations in lightning discharges were drawn from the measurements.

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## § 1. INTRODUCTION

FOR several years investigations have been carried out at Husbyborg, near Uppsala, in connexion with the electric properties of thunderstorms, and especially of the discharge characteristics of lightning. In the preliminary stages the work comprised measurements of the slow variation of the electrostatic field in thunderstorms. After some time the main interest turned to investigation of the rapid variations of the electromagnetic field produced by lightning discharges. Experiments in the first place were concentrated on the rapid variations of the electric field. Researches on this subject were especially desirable in regard to an important practical interest—the surges caused on high-tension transmission lines by lightning discharges.

With regard to such a practical application our investigations were in their first stage supported by the Royal Swedish Board of Waterfalls, an enterprise which is responsible for the high-tension transmission lines belonging to the State. A laboratory for surge phenomena and allied high-tension problems was consequently erected at Husbyborg. This laboratory became in 1932 incorporated with a newly erected research institute, which belonged to the University of Uppsala. The new institute had a special scientific purpose, being mainly devoted to the problems of high tensions, electric discharges in the atmosphere, and allied phenomena.

Researches on lightning discharges were thus obviously of special importance to the new institute, and the earlier investigations of the rapid variations in the electric field caused by lightning were continued. The results have been published elsewhere<sup>(1)</sup>. The relations between the variations in the electric field caused by lightning and radio atmospherics were analysed also<sup>(2)</sup>. In addition to these pro-

blems we have been occupied with an investigation of the rapid variations in the magnetic field caused by lightning discharges. The main purpose in this undertaking was to analyse the discharge variation of lightning, with special reference to the structure of the currents in the lightning paths. During the season of 1936 the experiments were continued, and improved recording methods, as compared with earlier investigations<sup>(3)</sup>, were used. A considerable amount of new data was collected and it will be the purpose of this paper to give an account of the researches and their main results.

## § 2. THE METHODS OF RECORDING

*The measuring instrument.* Earlier and preliminary experiments which indicated that the variations within a lightning flash are very rapid enabled us to decide upon the most suitable methods for recording lightning discharges. The total time of discharge in a lightning flash is, on the other hand, rather slow and very often exceeds several hundredths of a second in duration. A lightning discharge was found to be built up of sequences of individual discharges, which sometimes comprised time intervals as small as a few tenths of a microsecond. For instance an individual examination of the slope of the front of the pulse in such partial discharges required an accuracy of measurement of some tenths of a  $\mu$ sec. From this it was evident that the only instrument which could be used with any hope of success must be a cathode-ray oscillograph. Further, it was found that the time variations were so rapid that a cathode-ray tube of the sealed type had not a sufficiently high recording-velocity. Hence it was necessary to use an oscillograph in which the film was placed in a vacuum and directly bombarded by the electron beam from the cathode.

During the experiments a special difficulty arose. When the oscillograph was in operation to record a lightning discharge it was not possible to obtain beforehand any indication of the moment of occurrence of the discharge. Thus a lightning flash with a duration of e.g. 0.001 sec. would require an exposure time of the film up to 10 min. or more. The film in an ordinary oscillograph with such a length of exposure would be much over-exposed, and no rapid transient phenomena could be recorded or recognized on such a film.

A means of avoiding these difficulties would be an electron-tube relay, which would operate by the lightning discharge. The arrangement should operate so that the electron beam of the oscillograph had to be directed into a recording position by means of the relay. The discharge variation of a lightning flash showed, however, such complicated variation forms that the relay would necessarily be very complicated. It was further not very advisable to introduce in the observation circuit a relay with a more or less pronounced time lag. Such an arrangement would, for instance, necessitate propagation of the complicated discharge variation in a delay cable during the time necessary to operate the relay.

It was evident, from the very beginning, that the problem had to be solved in such a manner that the electron beam of the oscillograph itself should be made to function as a relay. By this method a special advantage was gained, which was most

useful in the recording of the complicated forms of variation in lightning discharges. With the relay the measuring circuit became very simple and easy to operate.

The principle which throughout has been used in the records of lightning discharges is illustrated in figure 1. The operation of the relay was as follows. When the plates 3, 5 have no voltage, the electron beam will pass from the cathode through the anode-hole between the plates 3 and against the target 4, which has such a position and dimensions that its shadow covers the central hole of the screen 6. A voltage variation derived from the potentiometer 9 and impressed upon the plates 3, 5 will bend the electron beam around the target, and by the cross-coupled system 5 the beam will be bent back and will pass through the hole 6. The plates 7 provide the time base from a circuit with either sinusoidal or linear

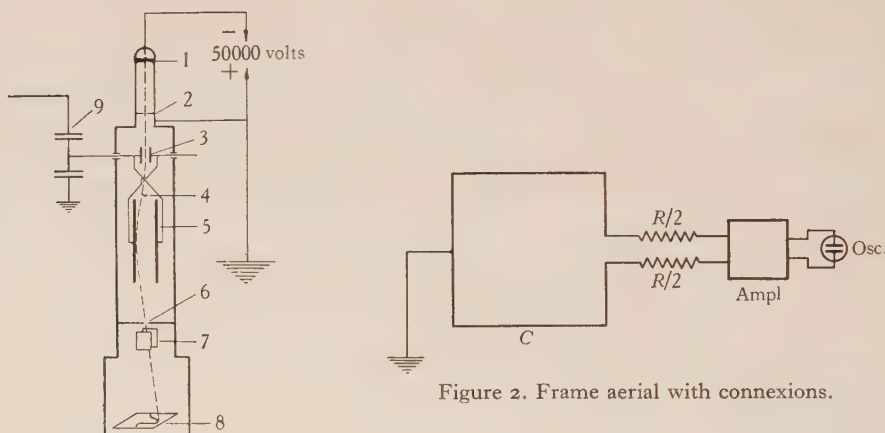


Figure 1. Relay of cathode-ray oscillograph. 1, cathode; 2, anode; 3, 5, deflection plates; 4, target; 6, screen with a central hole; 7, deflection plates for time circuit; 8, record film; 9, potentiometer.

time variation. The cathode-ray beam will thus fall upon the recording film only during the passage of the impulse, so that the target produces a characteristic zero line or band shadow in the oscillograms. This can be seen in the records here reproduced. In the majority of cases there is no difficulty in interpolating to determine the path of the beam across the zero band.

The relay method described by the author<sup>(4)</sup> has been used in various experiments in which the purpose was to record impulses whose time of arrival was not known beforehand. Typical of such unexpected impulses are lightning surges on transmission lines, impulses in radio antenna circuits, and atmospherics arriving from long distances. The method proved also to be very convenient in the recording of the rapid variations in the magnetic field.

The cathode-ray oscillograph constructed by the author and used in these experiments was of the cold-cathode type. The instrument is built of metal with the exception of the glass tube between the cathode and the anode.



The oscillograph contains a film-holder of such construction that the film can be moved to an exposure position from outside. It is possible to make up to 100 exposures without opening the oscillograph. The large number of exposures necessitated loading the film-holder with rolls of film of a length up to 10 metres. Such long films required a very safe and effective system of evacuation. The operation of the oscillographs had to be carried out in movable field stations without any supply of cooling-water. It was thus very inconvenient to use diffusion pumps. A mechanical molecular pump with a conical rotor was therefore constructed for the experiments. The use of a conical rotor had the advantage that the distance between the rotor and the cams between the spiral groove could easily be adjusted to give a very small clearance. The small distance allowed of a very high pump efficiency with as low a rotation speed as 3000 rev./min.

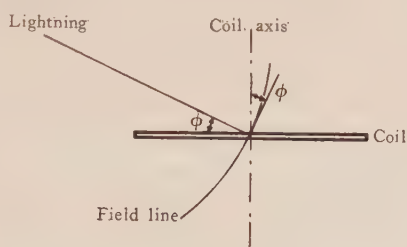


Figure 3. Orientation of coil to lightning path.

During the experiments it often happened that the time between lightning discharges was many minutes. The relay arrangement described did not fully prevent fogging of the film. After an exposure of some minutes it was necessary to shift to a fresh film-surface and thus it was possible that the supply of film in an oscillograph would be totally exposed during thunderstorms of a long duration. In order to avoid such a situation two oscillographs were simultaneously in operation. Both were charged with film and evacuated before the beginning of the observations. During the observation periods one instrument was used for the record and the other for indication by direct observation. The records were shifted over to the oscillograph used for indication when the film-supply of the first recording instrument was consumed, and the instrument with exposed film was subsequently used for indication. This alternation of the instruments was very useful and allowed of very long and undisturbed periods of operation.

### § 3. EXTERNAL EXPERIMENTAL ARRANGEMENTS

Frame aerials of special construction were found to be most suitable for measuring the rapid variation of the magnetic field. Certain precautions were necessary with regard to the winding and the dimensions of the coils; thus it was impossible to use coils with compact turns on account of pronounced disturbances arising in such a case between consecutive turns. It was necessary to separate the wire of each turn by an air distance of at least 2 cm. The coils showed with certain types of

discharge-variations a tendency to become the seat of resonant oscillations, which were eliminated by suitable damping resistances.

In our first experiments the ends of the coil were directly connected to the deflection system of the oscillograph. In such a case the dimensions of the coils were to some extent determined by the voltage-sensitivity of the oscillograph. With a distance between the observation station and the lightning path of from 2 to 5 km. we received voltage-variations up to 800 V., which was more than sufficient for a record on the instrument. The coils used in such a case had a turn-surface of  $1.95 \times 10^7$  turns per  $\text{cm}^2$  and the wire consisted of copper with a diameter of 1 mm. The connexion of the coil to the oscillograph was symmetrical and the mid-point of the coils was earthed as is illustrated in figure 2. The damping resistance  $R$  was  $6000 \Omega$ .

Direct control of the oscillograph from the coil was inconvenient as the limited sensitivity only permitted the stronger variations of the magnetic field to be recorded.

Therefore, as has already been mentioned, we introduced during the experiments of the summer 1936 an essential improvement in which the voltage-variations of the frame aeriels were amplified by means of a special arrangement. Between the end of the coil and the oscillograph was placed a voltage-amplifier with an amplification up to 800 times. A necessary condition was that this amplifier should work without distortion within the range of variation in the observations. Further, the arrangement had the advantage that smaller amplitude-variations in the field could be recorded, and we received thus more extensive experimental data. The dimensions of the frame aeriels were also considerably diminished by the introduction of an amplifier. The coils in the earlier arrangement had a surface of  $130 \text{ m}^2$ , the coils of the new arrangement had a surface of only  $12 \text{ m}^2$ . In the latter case the small dimensions made it possible to use a coil which could easily be turned around a vertical axis. The large coils used in our earlier experiments necessitated, on the other hand, two separate coils mounted in two vertical planes at right angles.

#### § 4. THE PRINCIPLE OF THE MEASUREMENTS

The observation of the magnetic-field variations was carried out by using the well-known induced effect of the magnetic fields on frame aeriels. We introduce the following symbols:  $H$  is the horizontal component of the field in c.g.s. units,  $A$  is the surface of the frame in  $\text{cm}^2$ ,  $n$  is its turn number,  $V$  is the induced voltage in volts, and  $\phi$  is in relation to figure 3 the angle between the coil and the direction of the lightning. The relation (1) will be as follows:

$$V = nA \frac{dH}{dt} 10^{-8} \cos \phi \quad \dots\dots(1).$$

It is obvious that the horizontal component of the field force  $H$  can be calculated from equation (1) by integration, and

$$H = \frac{10^8}{nA \cos \phi} \int V dt \quad \dots\dots(2).$$

The values of  $H$  in what follows have been calculated from this relation.

### § 5. THE CALCULATION OF THE LIGHTNING CURRENTS

Calculation of the current-variation in a lightning flash from the variation of the magnetic field would require a knowledge of the manner in which the magnetic field varied with the distance from the path of the lightning. The ideal solution of this problem would have been to use simultaneous observations from stations situated at suitable distances from the path. Such a method would allow of a determination of the magnetic moment of the lightning discharges; it has not yet been used, however, because we found it to be too expensive since it involved duplication of the experimental arrangements.

Under certain conditions, as has already been shown in my earlier publication on lightning currents, it was possible to use an approximation method in the calculation of the current variations. The calculations were restricted to vertical lightning paths and the following relation was used:

$$I = 10 V \frac{H \sqrt{(r^2 + h^2)}}{h} \quad \dots\dots(3),$$

where  $I$  is the current in the lightning path,  $H$  the horizontal component of the magnetic field,  $r$  the distance of the lightning path from the observation station and  $h$  the mean vertical length of the lightning path. For this mean height we have taken a value of 2.5 km. This was a plausible mean value in accordance with other observations which we have carried out on such heights. The amplification of the coil voltages resulted in an increase of several kilometres in the distance between the observation point and the lightning path. The uncertainty of  $\pm 0.5$  km. in the height of the lightning path will for such longer distances cause an error in the calculated current-values of about  $\pm 12$  per cent. The errors are smoothed out in the extended series of observations given in this paper.

The displacement currents may influence the determinations of the current-values of the lightning path, especially for the longer distances and to an extent, for the most disadvantageous cases, which has been estimated at 10 per cent. We have further supposed that the magnetic field lines will follow in concentric circles around the lightning path. Topographic conditions can in this respect provoke certain local disturbances. In our case no such disturbances have influenced the results. We estimate the total probable errors in the calculated values of the lightning currents under the most unfavourable conditions to be 20 to 22 per cent. Such errors must on the other hand be smoothed out to a great extent in an extended series of observations. As will be seen from what follows, the individual values of the maximum lightning currents change from a thousand to a hundred thousand amperes, and an error in the individual determinations will thus not be of any appreciable importance when we consider the problem as a whole.

### § 6. OBSERVATIONS

When a thunderstorm passed near the observation station and was suitably located for the production of a record, the frame aerial was directed in accordance with the most probable direction of the lightning paths. One or two observers



noted the actual paths of the lightning with reference to their azimuthal positions and to previously determined markings on the horizon. The method provided a sufficiently accurate determination of the direction of the lightning tracks. These were also noted as carefully as possible as regards their angular size and general aspect. The distance to the flash was determined by the time difference between lightning and thunder.

A calculation of the maximum current-variation could be carried out only for vertical lightning flashes. A more limited analysis, however, of discharge characteristics, for instance the time sequences of consecutive partial discharges, and the total duration of lightning discharges were taken also from lightning paths inclined to the vertical or observed to be horizontal.

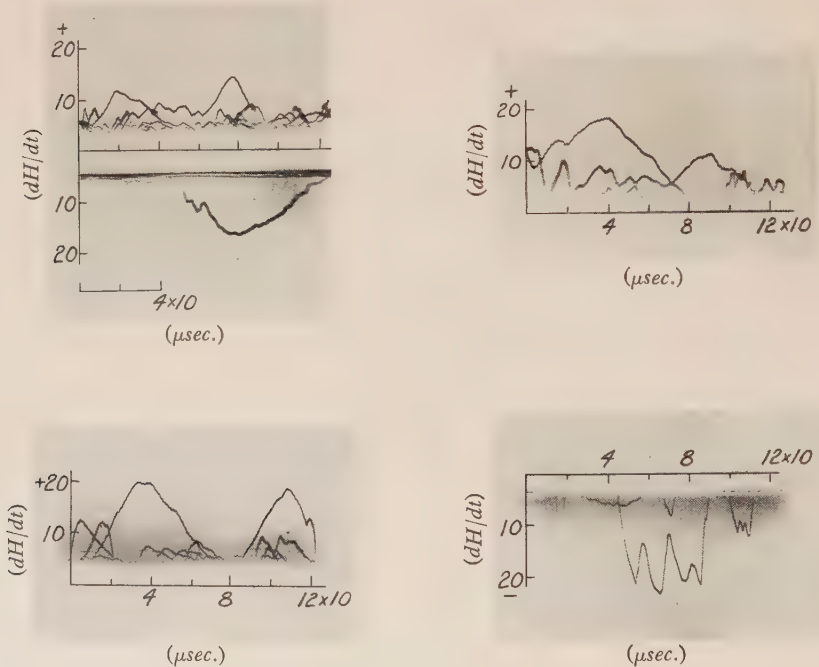


Figure 4. Oscillogram taken with frame aerial, with rapid base time.

Earlier experiments relating to the rapid variations of the electric field caused by lightning proved that, with few exceptions, an ordinary discharge visible as lightning is composed of consecutive partial discharges. The total duration reached values up to a hundredth part of a second. The partial discharges were on the other hand sometimes as short as only  $10\mu\text{sec}$ . If the oscillograph had its time-base circuit adapted to such short time variations, very complicated oscillograms were obtained from which it was necessary to deduce the individual variations. An example of such complicated oscillograms is reproduced in figure 4. In order to obtain a more general analysis of the variations it was also necessary to operate

with a much slower time base. Oscillograms of this kind were better adapted to an analysis of consecutive partial discharges. Such oscillograms are exemplified in figure 5.

#### § 7. DISCUSSION OF THE RESULTS

As has already been mentioned, our first measurements of the variation in the magnetic field were carried out without an amplifier. The advantage of this direct method was that it records where very rapid variations could be obtained without any distortion; its disadvantage was the limitation of sensitivity, and consequent restriction of the observational data which it involved.

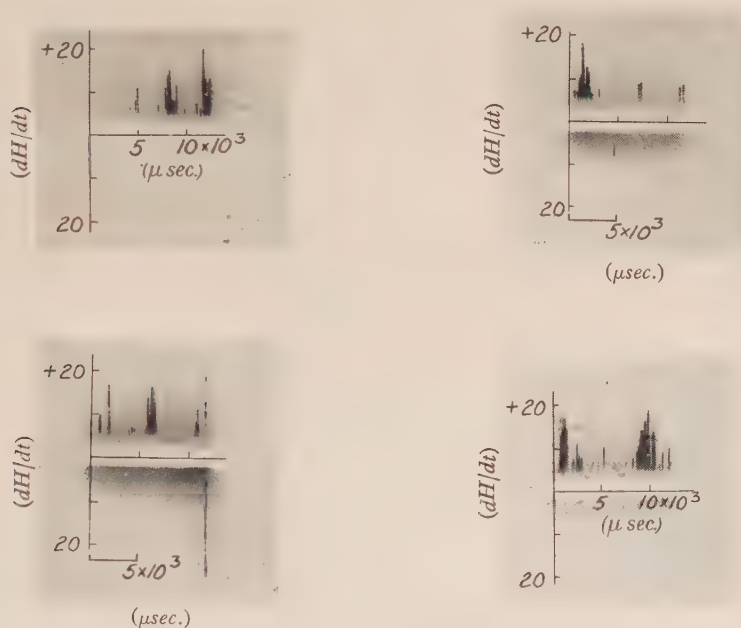


Figure 5. Oscillogram taken with frame aerial, with slow base time.

The advantage of the method now described was that it yielded a substantial increase in the number of observations within a relatively short observation period. On the other hand the improved method was not without inconveniences. With the increase of the distance between the place of observation and the lightning path a transformation in the form of the impulses must ensue. This was especially the case for the variation in the electric field. The influence was not so pronounced for the corresponding variation in the magnetic field and could be completely neglected in the case of the values given in this paper.

The distances between the lightning paths and the point of observation were sometimes so great in the present experiments as to render it possible that a damping effect might be influencing the results. The result of this damping must in the first

place have been a slight alteration of the steepness of the front of the impulse in such a way that the time necessary for the current to reach its peak value became longer. A slight indication of such damping effects is visible in some of our records with the longest distances to the lightning path.

#### § 8. THE CHARACTERISTIC PROPERTIES OF INDIVIDUAL CURRENT PULSES

The oscillograms provided data for a calculation of the total duration of the current pulses in fifty lightning discharges. The result is reproduced in figure 6, from which it is seen that the majority of the discharges had a duration below 0.005 sec.

The time intervals between successive partial lightning-current pulses for a number of 344 individual cases are reproduced in figure 7. The most usual time interval is characterized by values below 0.001 sec.

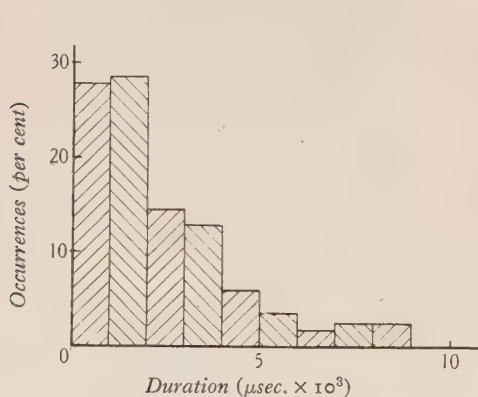


Figure 6. Total duration of current pulses.

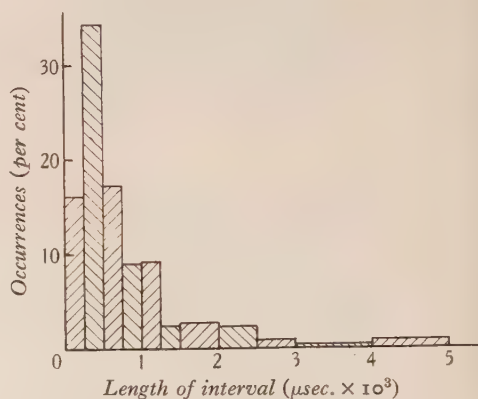


Figure 7. Time intervals between successive partial discharges.

The percentage distribution of the number of current pulses in 127 lightning discharges is reproduced in figure 8, from which it follows that the most usual number consisted of three separate pulses.

It is of interest to analyse the distribution of the maximum current amplitudes in successive individual pulses. In lightning discharges consisting of four or more current pulses the maximum current amplitudes were found in 85 per cent of the cases to occur in the two first pulses, or within the first half of the discharge. In the case of lightning discharges consisting of three pulses the maximum current was found quite as often in the first as in the second of the three discharges.

The percentage distribution of the total duration of individual pulses is reproduced in figure 9. The most usual values reached durations of from 20 to 60 μsec. In our first investigation the corresponding values were from 10 to 30 μsec. This difference in values can be fully explained by the difference in sensitivity during the



two periods of observation. In our latest experiments we have used an especially sensitive method, by which impulses with low amplitudes and longer durations also could be recorded.

In 364 discharges we have calculated the time for the current to reach its crest value, and the results are reproduced in figure 10. The most usual values were from 6 to 8 or from 4 to 6  $\mu\text{sec.}$ , while earlier observations resulted in corresponding values of from 3 to 6  $\mu\text{sec.}$

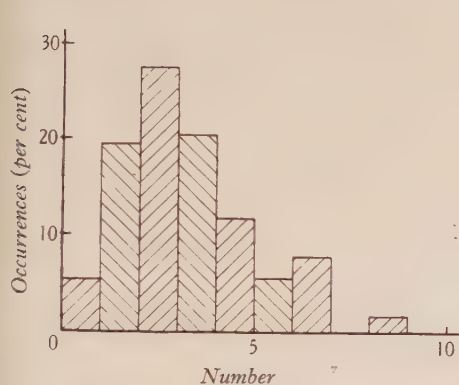


Figure 8. Number of partial discharges.

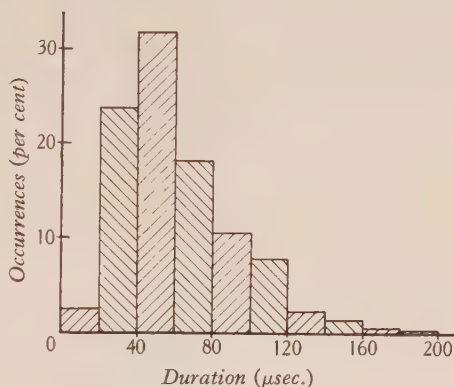


Figure 9. Total duration of individual pulses.

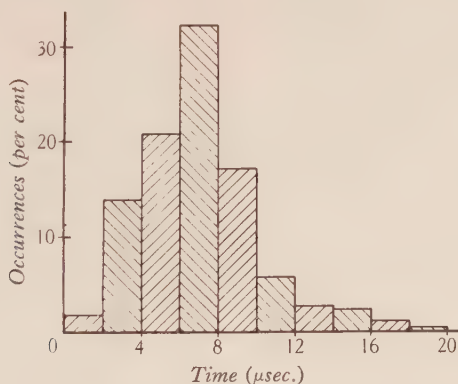


Figure 10. Time intervals for peak values of lightning current.

#### § 9. CALCULATED HORIZONTAL COMPONENTS OF THE MAGNETIC FIELD

The horizontal components of the magnetic field in c.g.s. units have been calculated from observed vertical lightning discharges and are reproduced in figure 11. The positive sign is used to indicate that a positive charge goes to earth. Of eighty-eight recorded vertical lightning discharges 47 per cent were positive and 53 per cent negative, in full accordance with earlier results.

## § 10. CALCULATION OF THE MAXIMUM CURRENT-VALUES

During the observation period, vertical lightning discharges were in twenty-eight cases so located with regard to the observation station that it was possible to use the relation (3) for calculating the maximum current-values. The observations allowed of calculation for sixty-five partial discharges within the observed lightning paths, and the distribution of the values is shown in figure 12. The data are in full agreement with our earlier results, where the maximum amplitude reached was 120 kA. Figure 12 shows a considerable number of values as low as 1–5 or 6–10 kA. This occurrence of small currents is explained by the improvement in the sensitivity of the experimental methods.

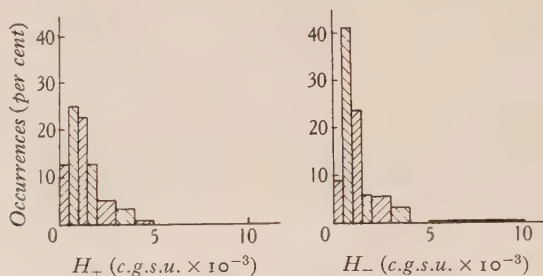


Figure 11. Horizontal components of magnetic field.

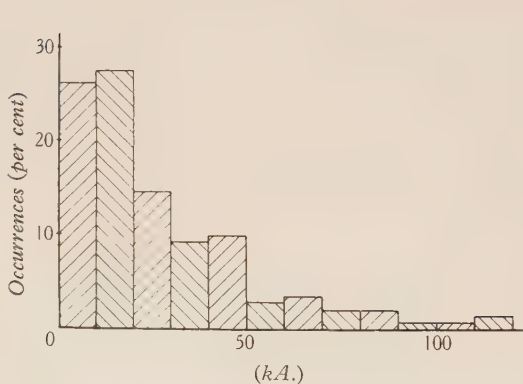


Figure 12. Distribution of peak values of individual current pulses.

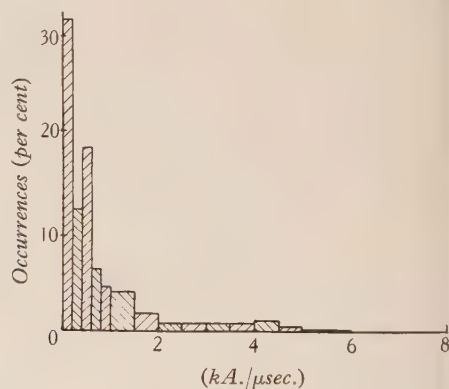


Figure 13. Maximal changes with time in lightning currents.

It is of special interest to compare the calculated maximum current-values with results from an independent and indirect method. On a large number of steel towers supporting high-tension power lines in the United States<sup>(5)</sup> and Germany<sup>(6)</sup>, small steel links have been used to observe the maximum currents of lightning discharges which strike the towers. The steel of the link consists of a cobalt-nickel-iron alloy or a coal steel with high magnetic retentivity. A lightning stroke to a tower fitted with such a link resulted in magnetization of the link by the current.

The values of the magnetization could be determined subsequently, and from a knowledge of the magnetic constants and the location of the link on the tower it was possible to calculate the current passing through the tower. About 4000 observation stations were used in the United States for this purpose during the years 1932 to 1936.

The maximum observed value was 220·000 A. and there were relatively few values exceeding 100·000 A. The majority of the currents did not attain such high maximum values as 100·000 A. and were in very good agreement with our own values as regards their general distribution. The observational data from Germany were not so extensive as the data from the United States, but they also were in good agreement with our own results. An evident difference existed between our observations and those mentioned above. Both the American and the German observations resulted in a marked preponderance of negative currents, while our measurements indicated only a slight preponderance of this polarity. This smaller preponderance of negative values was quite in agreement with other and earlier observations of the polarity of the rapid variation in the electric field. It must be pointed out that the measurements made by the link method were carried out on very well earthed steel towers. Our measurements on the other hand were carried out in the open field. The marked preponderance of negative current-values for such structures as steel towers must, as we have found, be caused to a considerable extent by a polarity effect. Thus under the same conditions of field-intensity and charge-distribution, lightning discharges over steel structures will be much more easily developed at lower voltages when the cloud-polarity is negative than when it is positive.

From our observations it was possible to calculate the maximum changes of current with time in the lightning currents shown in figure 12. The result is graphed in figure 13. In our earlier investigations we obtained a few values of maximum current-changes amounting to 24–30 kA./ $\mu$ sec. These values were considered to be exceptional and have not recurred in the more recent work. Figure 13 shows also a preponderance of small changes of current with time, and this is to be expected with the improved sensitivity of our experimental methods.

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# THE KNIFE-EDGE TEST FOR SPHERICAL ABERRATION

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**ABSTRACT.** A method is given for computing the appearance seen in the knife-edge test, for various positions of the edge in various focal planes, when any specified amount of aberration is present. Series of patterns are given corresponding to (a) pure primary spherical aberration, and (b) zonal aberration with marginal correction.

## § 1. INTRODUCTION

THE knife-edge test as originally described by Foucault<sup>(1)</sup> was used as a means of ascertaining whether or not a telescope objective was perfect, and also as a guide in the improvement of an objective by local retouching. However, the knife-edge method may also be very usefully applied to lenses having considerable amounts of aberration, as has been shown by Miss Conrady<sup>(2)</sup>.

It was felt desirable to extend this work, particularly in respect of spherical aberration. It will be realized that there is really a double infinitude of possible knife-edge shadows for a given lens, since the knife blade may be situated in any focal plane, and the edge itself may be advanced to any distance into the beam in that plane. In Miss Conrady's paper, only one shadow appearance is given for spherical aberration, namely that for the mid-position of the knife edge in a focal plane about midway between the marginal and paraxial foci.

The aim of the present paper is to explain a method by which the form of the shadow edge for any mixture of aberrations, at any specified position of the knife edge, may be computed.

## § 2. THEORY OF THE METHOD

It will be clear from elementary considerations that, if a lens possesses a considerable amount of spherical aberration, any zone of the lens will suddenly go dark as the knife edge cuts into its exact focus. Other zones of the lens, of which the foci lie beyond the knife plane, will darken on the side on which the knife is entering, whereas zones whose foci lie closer to the lens than this plane will darken on the opposite side. The result will be an appearance shown diagrammatically in figure 1.

The accurate computation of this and other shadow appearances can be made by differentiating the ordinary expression for spherical aberration as a path difference, namely:

$$P = \frac{A}{2l} r^2 + \frac{B}{4l} r^4 + \frac{C}{6l} r^6 + \dots \quad \dots\dots(1).$$

Here  $A$  represents a change of focus from the paraxial focal plane;  $B$ ,  $C$ , etc. represent successive orders of spherical aberration;  $r$  is the radius of the zone; and  $l$  represents the distance of the image from the lens.

If we let  $(x, y)$  represent the two principal meridians of the lens, respectively perpendicular and parallel to the knife edge, then the path-difference may be written:

$$P = \frac{A}{2l}(x^2 + y^2) + \frac{B}{4l}(x^2 + y^2)^2 + \frac{C}{6l}(x^2 + y^2)^3 + \dots \quad \dots\dots(2).$$

By differentiating this, the components of the slope  $\alpha$  of the ray from the point  $(x, y)$ , relative to the ideal path of that ray, are obtained:

$$\left. \begin{aligned} \alpha_x &= \frac{A}{l}x + \frac{B}{l}x(x^2 + y^2) + \frac{C}{l}x(x^2 + y^2)^2 + \dots \\ \alpha_y &= \frac{A}{l}y + \frac{B}{l}y(x^2 + y^2) + \frac{C}{l}y(x^2 + y^2)^2 + \dots \end{aligned} \right\} \quad \dots\dots(3).$$

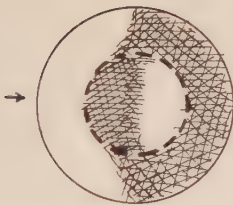


Figure 1. Knife-edge shadow, when knife blade is at focus of dotted zone (schematic).

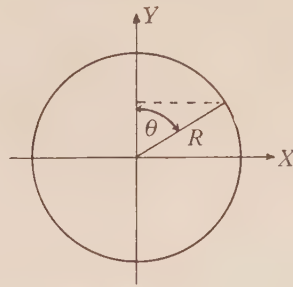


Figure 2. The principal meridians on the lens aperture.

These may be simplified by using polar coordinates on the lens aperture, figure 2, thus

$$\left. \begin{aligned} x &= r \sin \theta \\ y &= r \cos \theta \end{aligned} \right\} \quad \dots\dots(4).$$

We now convert equation (3) to give the linear-displacement components  $\xi$ ,  $\eta$  of the point at which the ray intersects the chosen focal plane. Since  $\xi = -l\alpha_x$ ,  $\eta = -l\alpha_y$ , we have:

$$\left. \begin{aligned} \xi &= -\sin \theta [Ar + Br^3 + Cr^5 + \dots] = -K \sin \theta \\ \eta &= -\cos \theta [Ar + Br^3 + Cr^5 + \dots] = -K \cos \theta \end{aligned} \right\} \quad \dots\dots(5).$$

The procedure for computing the knife-edge patterns is to calculate the values of  $K$  for a series of values of  $r$ , say  $r = 0.1, 0.2, 0.3 \dots 1.0$ , and then tabulate the corresponding values of  $\sin \theta$ , equal to  $-\xi/K$ , for each of a series of values of  $\xi$ . The values of  $\theta$  itself are then tabulated for all values of  $\sin \theta$  less than unity. Now since each value of  $\xi$  represents a definite stage in the progress of the knife edge across the beam, the polar graph of  $(r, \theta)$  for each value of  $\xi$  gives the appearance of the shadow edge for that position of the knife blade.

§3. THE PATTERNS FOR PRIMARY SPHERICAL ABERRATION

As an example, suppose it is desired to compute the shadows for pure primary spherical aberration, the knife blade being supposed to lie in the marginal focal plane. Then if  $B=1$  (say),  $A$  will be  $-1$ , if  $r$  is taken as unity at the margin of the lens. Thus  $K=r^3-r$  in this case. The values, with reversed sign, of  $\sin \theta$  and  $\theta$  to the nearest degree, corresponding to a series of values of  $\xi$ , are shown in table 1.

Table 1

Values of  $-\sin \theta$  and  $-\theta$  (degrees) corresponding to given values of  $r$  and  $\xi$ . The values of  $\theta$  are printed below those of  $\sin \theta$ .

$r \backslash -\xi$	$\cdot 05$	$\cdot 1$	$\cdot 15$	$\cdot 2$	$\cdot 3$	$\cdot 4$	$\cdot 5$	$\cdot 6$	$\cdot 7$	$\cdot 8$	$\cdot 9$	$1$
$-K$	$\cdot 050$	$\cdot 099$	$\cdot 147$	$\cdot 192$	$\cdot 273$	$\cdot 336$	$\cdot 375$	$\cdot 384$	$\cdot 357$	$\cdot 288$	$\cdot 171$	$0$
$\cdot 05$	$1\cdot 002$ —	$\cdot 505$ 30	$\cdot 344$ 20	$\cdot 265$ 15	$\cdot 183$ 11	$\cdot 149$ 9	$\cdot 133$ 8	$\cdot 130$ 7	$\cdot 140$ 8	$\cdot 173$ 10	$\cdot 292$ 17	$\infty$ —
$\cdot 10$	$2\cdot 0$ —	$1\cdot 01$ —	$\cdot 688$ 43	$\cdot 521$ 32	$\cdot 366$ 21	$\cdot 298$ 17	$\cdot 267$ 15	$\cdot 260$ 15	$\cdot 280$ 16	$\cdot 347$ 20	$\cdot 585$ 36	— —
$\cdot 15$	— —	— —	— —	$\cdot 786$ 52	$\cdot 549$ 33	$\cdot 447$ 26	$\cdot 400$ 24	$\cdot 390$ 23	$\cdot 420$ 25	$\cdot 520$ 32	$\cdot 877$ 61	— —
$\cdot 20$	— —	— —	— —	— —	$\cdot 732$ 47	$\cdot 596$ 36	$\cdot 534$ 32	$\cdot 520$ 31	$\cdot 560$ 34	$\cdot 694$ 44	— —	— —
$\cdot 30$	— —	— —	— —	— —	— —	$\cdot 894$ 63	$\cdot 801$ 54	$\cdot 780$ 53	$\cdot 840$ 58	— —	— —	— —
$\cdot 40$	— —	— —	— —	— —	— —	— —	— —	— —	— —	— —	— —	— —

These values are most conveniently plotted on ordinary polar-curve paper, a circle of ten units diameter being taken to represent the rim of the lens aperture. Graph points are marked on this corresponding to all the available pairs of values of  $r$  and  $\theta$ , figure 3. The locus of all points having the same value of  $\xi$  represents the

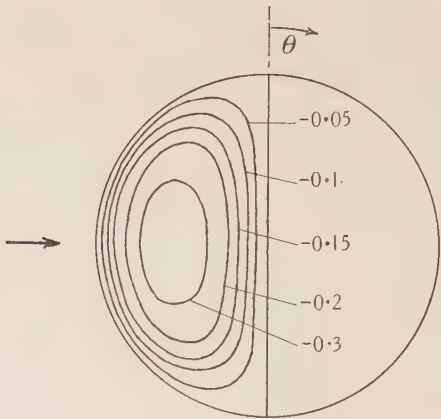


Figure 3. Computed shadow edges for primary spherical aberration, in marginal focal plane.



edge of the shadow corresponding to a knife-position given by  $\xi$ ; thus by this method the whole family of shadow-boundaries are plotted on the same figure. They can be then traced off in turn and collected as shown in figure 4. For the different focal planes in the diagram, the values of  $A$  were respectively  $-1.5$ ,  $-1.0$ ,  $-0.5$ ,  $0$ , and  $0.5$ .

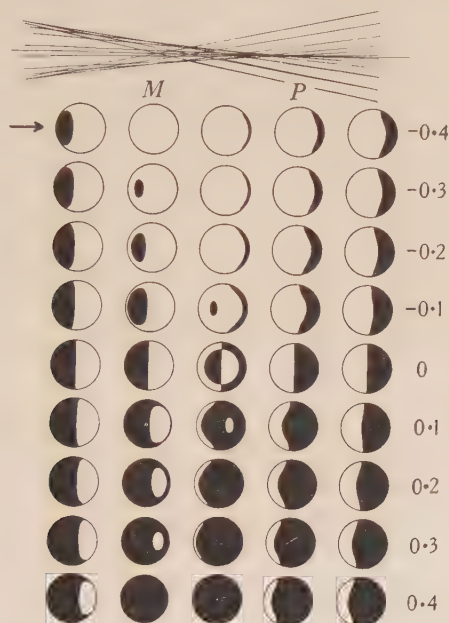


Figure 4. The knife-edge shadows for primary spherical aberration (computed).

In figure 5 is shown a series of actual photographs of knife-edge shadows made on a reversed telescope objective, the position of the knife blade being removed progressively further from the lens from left to right. The knife edge was advanced in each case from above to a point as near to the lens axis as could be secured.



Figure 5. Knife-edge shadow photographs for a reversed telescope objective.

One very characteristic feature of this test is the appearance seen when the knife blade is at the paraxial focus. The central patch of shadow is seen to jump suddenly across the centre of the aperture for an infinitesimal advance of the knife blade. This appears to be the only method of experimentally locating a paraxial image point, since it does not require the stopping down of the lens to a small hole with the consequent great increase in the depth of focus.

## §4. THE PATTERNS FOR ZONAL ABERRATION

In the case of zonal aberration, since the marginal aberration is to be zero, the values taken were  $B = +1$ ,  $C = -1$ ,  $A$  having the values  $-0.375$ ,  $-0.25$ ,  $-0.125$ ,  $0$ ,  $+0.125$  respectively, and  $\xi$  the values  $-0.15$ ,  $-0.10$ ,  $-0.05$ ,  $-0.01$ , and  $0$ .

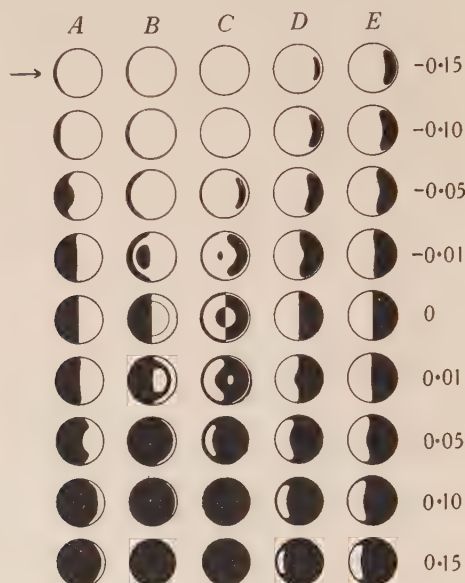


Figure 6. The computed knife-edge shadows for zonal aberration.

The plane for which  $A = -0.25$  (column B, figure 6) is the plane of the minimum zonal focus, namely the focus of the zone with  $r$  equal to  $0.707$ . Column D corresponds to the united marginal and paraxial focus, in which plane the coefficient  $A = 0$ .

## §5. CONCLUSION

In the discussion given above, only the first two orders of spherical aberration have been considered; the same method is of course applicable to the computation of knife-edge shadows for any single aberration or mixture of aberrations which is expressible as a path difference in equation 1. When aberrations such as coma or astigmatism, which do not have rotation symmetry about the lens axis, are present, it is clear that different shadow patterns will be obtained for different directions of entry of the knife blade. Some examples of these are given by Miss Conrady in the paper cited.

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# THE VELOCITY-DISTRIBUTION IN A LIQUID-INTO-LIQUID JET

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**ABSTRACT.** The distribution of velocity in a liquid-into-liquid jet, issuing from a cylindrical tube with a tapering approach, has been measured by photographing the traces made by suspended particles, with strong illumination and a determined exposure. The results have been compared with the theory for a jet issuing from a point in an infinite plane, and it is shown that, supposing the equivalent point source to be within the orifice, good agreement is found except in the immediate neighbourhood of the orifice. An expression has been derived connecting the distance of the equivalent point source from the orifice with the Reynolds's number. This enables the loss of kinetic energy of the jet to be worked out.

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## § 1. INTRODUCTION

FOR some time experiments have been in progress in the physics laboratory at University College, London, on the instability of a liquid-into-liquid jet. In particular, attention has been devoted to the nature of the motion produced by a periodic disturbance, acting on a fine jet of water flowing through a circular orifice into a shallow tank of water. In this connexion it is of interest to determine the distribution of velocity in the undisturbed jet, especially as comparatively little has been done experimentally on streaming without rigid walls.

The behaviour of a laminar jet of viscous liquid has been recently worked out by Schlichting<sup>(1)</sup> on certain simplifying assumptions, of which the most serious is that the jet issues from a point aperture, giving, as it does, an infinite velocity. One of the objects of this work was, therefore, to see how far Schlichting's theory could be applied to an actual jet, issuing from a circular aperture of finite size. The theory has proved to be very successful in interpreting the results, and it is shown here how, with slight modification, it can be made to give the velocity everywhere except in the immediate neighbourhood of the orifice. One consequence of this work is that the loss of kinetic energy due to viscosity can be worked out.



## § 2. EXPERIMENTAL METHOD

The experiments, being carried out in a physical laboratory, had to be done on a small scale. The internal profile of the glass jet used is given in figure 2, which shows a photograph obtained by immersing the glass jet, full of air, in a liquid of the same refractive index as the glass, contained in a glass tank with plane sides. The diameter of the orifice was 0.91 mm. The water used contained very fine suspended particles. A horizontal plane through the axis of the jet was strongly illuminated, and photographed with a measured short exposure. The tracks of particles, both in the water constituting the original jet and in the surrounding liquid, appeared as short lines terminating sharply, the lengths giving a measure of the velocity at the mid-point of the line. From these tracks, then, the velocity distribution could be obtained. Subsidiary experiments gave the volume per second passing through the jet at various pressures, measured in the straight approach tube. The pressure in this tube was used to control and determine the mean velocity of efflux.

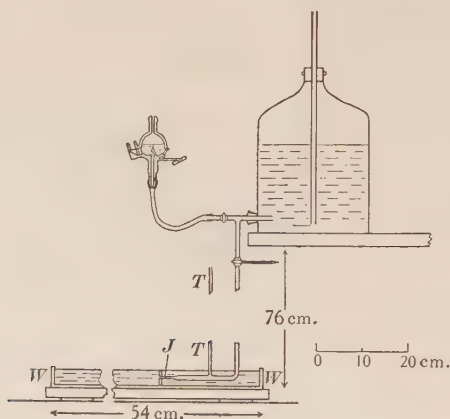


Figure 1.

The general arrangement of apparatus is shown in figure 1. Water from a large Mariotte's bottle, containing about 7 litres, was admitted, by a tap capable of accurate adjustment, to a vertical tube, leading to the jet *J*. The vertical tube *TT* allowed the pressure to be read by a cathetometer. The tank *WW* rested on a massive slate slab, supported on folded cloths, to minimize vibration. The level in the Mariotte's bottle was measured on a Casella sensitive pressure gauge, based on the internal reflection of a pointer from the surface of the water, reading to 0.01 mm. This was used to calibrate the outflow against the pressure, the time for the level to fall through 2 mm. being taken on a stop watch, and the corresponding volume being determined independently. The temperature was always maintained at 18° C., so that changes of viscosity had not to be considered.

Various particles were tried, including fish scales (as used for the manufacture of artificial pearls), flue dust and a powdered magnesium-aluminium alloy. Aluminium particles from commercial aluminium powder were finally adopted. The





Figure 2. (Magnification  $\times 1.5$ .)



Figure 3a. (Magnification  $\times 9.4$ .)



Figure 3b. (Magnification  $\times 9.2$ .)



powder was washed in a separating funnel with alcohol, to remove grease, and the coarser particles run off. The alcohol was then filtered off from the remaining particles, which were dried and then suspended in distilled water, in a vessel about 5 ft. high. The suspension was allowed to stand for about 15 min., and the middle part of the liquid column then drawn off. This liquid, diluted with a large bulk of water, was used for the experiments, being placed both in the tank and in the supply bottle. The particles were from 0.05 to 0.08 mm. in diameter.

For illumination, a 55 A. arc was focused on to a horizontal slit by a wide-angle condensing lens, and an image of the slit formed on the axis of the jet stream by a  $1\frac{1}{2}$  in. Mayer lens of aperture  $f:1.5$ . This image was about 0.1 mm. wide, so that, even allowing for the convergence and divergence of the beam away from the axis, only a central section of the jet was illuminated. To photograph the tracks a similar lens, by Dallmeyer, was used, which gave a magnification of 5 with the distances adopted. The plates were Ilford hypersensitive panchromatic, speed 8000 H. and D. to  $\frac{1}{2}$ -watt illumination.

A shutter was required which would give exposures of some thousandths of a second with an accuracy of 1 per cent or better. This was provided by a rectangular aperture in a weighted board, falling freely in its own plane under gravity; the horizontal edges of the aperture were accurately straight, and their separation was 2.3 cm. in the normal case, giving an exposure of 0.00782 sec. Another aperture often used gave an exposure of 0.00497 sec. The shutter fell within 2 mm. of the illuminating slit, and was controlled by a magnetic release.

The plates obtained gave sharply terminated lines, as exemplified in figures 3 *a* and 3 *b*, which are selected as showing typical results at two different velocities. To measure up a plate a reference line, drawn parallel to the axis of flow, was taken as a provisional  $x$  axis, the  $y$  axis being the normal to the axis through the orifice. The length of track was recorded against the coordinates of the mid-point of the track, several photographs being taken for each rate of efflux, so as to provide sufficient material. The exact position of the  $x$  axis was then obtained from considerations of symmetry. A standard value of  $x$  being selected, the velocity at this distance was obtained, if necessary by interpolation, for different values of  $y$ . Three different values of  $x$  were used, viz. 0.8, 1.8 and 3.3 cm.

### § 3. TREATMENT OF RESULTS

Schlichting's treatment, which is based on the neglect of the radial accelerations, in the sense of the boundary layer theory, leads to expressions for  $u$  and  $v$ , the longitudinal and radial velocities, of the following form:

$$u = \frac{3}{8\pi} \frac{K}{\nu x} \frac{1}{(1 + \xi^2/4)^2}, \quad v = \frac{1}{4} \sqrt{\frac{3K}{\pi}} \cdot \frac{1}{x} \cdot \frac{\xi - \xi^3/4}{(1 + \xi^2/4)^2} \quad \dots\dots(1),$$

where  $\xi = \sqrt{\frac{3K}{16\pi}} \cdot \frac{1}{\nu} \cdot \frac{y}{x}$  and  $K = \frac{J}{\rho}$ ,

$J$  being the momentum crossing a plane normal to the axis of the jet per second,  $\nu$  the kinematic viscosity and  $\rho$  the density of the fluid. The distance  $x$  is measured

parallel to the axis, from the point origin of the jet. The jet draws the surrounding liquid along, the theoretical stream lines in a particular case being given in figure 9.

We can express  $u$  in the following form:

$$u = A \frac{1}{(1 + By^2)^2} \quad \dots\dots(2),^*$$

where

$$A = \frac{3K}{8\pi\nu} \frac{1}{x}, \quad B = \frac{3K}{64\pi\nu^2} \frac{1}{x^2},$$

so that

$$A/B = 8\nu x.$$

If then we plot the experimental values of  $1/\sqrt{u}$  against  $y^2$  we should obtain a straight line, and this expectation has been realized. The intercept on the vertical axis gives  $1/\sqrt{A}$ , and the slope of the line gives  $B/\sqrt{A}$ , from which  $A$  and  $B$  can be calculated. With the values of  $A$  and  $B$  so found theoretical curves of  $u$  against  $y$  have been drawn for comparison with the experimental points.

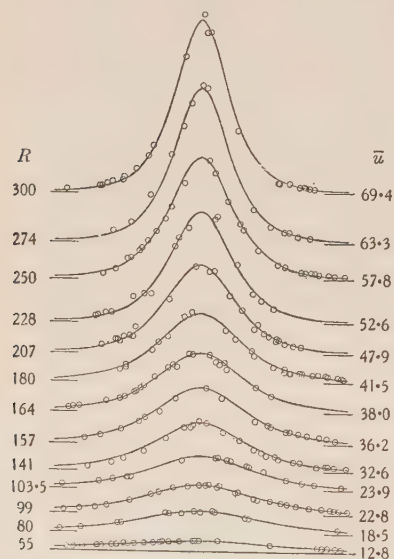


Figure 4.

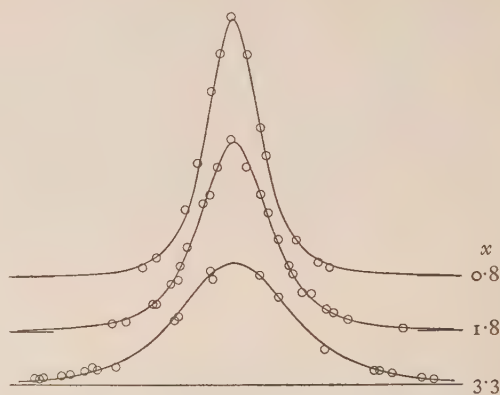


Figure 5.

Figure 4 shows the theoretical curves of velocity-distribution across the jet, and the experimental points, at a distance  $x = 3.3$  cm., for various mean velocities of flow in the jet itself, calculated from the volume efflux per second  $Q$  and the cross-sectional area of the jet, which was  $0.650 \text{ mm}^2$ . Actually the pressure  $p$  was measured in each experiment, and  $Q$  found from the experimental curve connecting  $Q$  and  $p$ . It will be seen that over a wide range (Reynolds's number in the tube, taken as  $R = \bar{u}a/\nu$ , where  $a$  is the radius of the orifice, varies from 55 to 300) the agreement between the theoretical and experimental forms is excellent. The results at three different distances from the jet, viz.  $x = 0.8, 1.8, 3.3$  cm., with a fixed value of  $\bar{u} = 46.7 \text{ cm./sec.}$ , are shown in figure 5, where the agreement is equally satisfactory.

#### §4. THE MOMENTUM OF THE JET

Velocity-distributions were measured at each of the known distances just specified, with various values of  $\bar{u}$ , and in this way a series of values of  $A$  and  $B$  were obtained. Now according to theory,

$$A/B = 8\nu x \quad \dots\dots(3),$$

and

$$K = \frac{\pi}{3} \frac{A^2}{B} = 1.047 \frac{A^2}{B} = \frac{J}{\rho} \quad \dots\dots(4),$$

$$= J \text{ in the case of water.}$$

Equation (4) enables us to find a value for  $J$  from each velocity profile measured. These various values are shown in figure 6, plotted against the pressure in centimetres of water, and it will be seen that the measurements made at three different distances are consistent.

The volume issuing from the jet per second has been measured in terms of the pressure, but to find the momentum we must know the velocity-distribution at the orifice, for which we have to consider whether, with the short tube used, the influence of the entry extended to the orifice. At first sight it would seem that we have

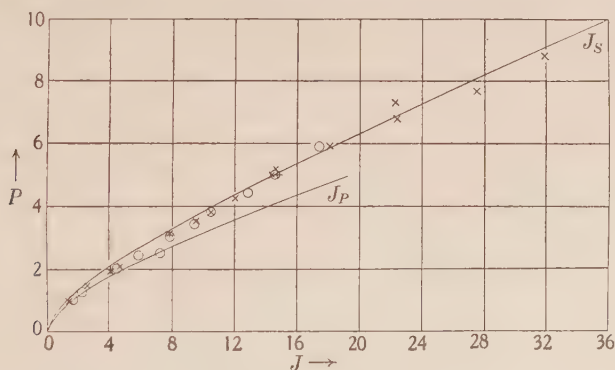


Figure 6.  $\times$ ,  $x=3.3$  cm.;  $O$ ,  $x=1.8$  cm.;  $+$ ,  $x=0.8$  cm.

as a guide the work of Schiller<sup>(2)</sup>, who finds that the Poiseuille regime sets in at a distance  $l$  from the entry given by

$$l/d = 0.029 R.$$

Taking for  $l$ , the effective length of our tube, 1.2 cm., which corresponds to the length  $AB$  in figure 2, and a mean diameter of 0.103 cm., we find that  $R$  must not exceed 400, which would indicate that the parabolic distribution is only beginning to fail at our highest speeds. However, it has been shown that even a slight convergence in a tube (1 mm. in radius on a length of 1 m., while  $R=1000$ ) causes a marked flattening in the velocity-distribution<sup>(3)\*</sup> while, for the case of convergent parallel walls (two-dimensional streaming) Hamel<sup>(4, 5)</sup> has shown that  $U$  deviates markedly from  $U_{\max}$  only at the walls, the velocity being uniform across almost the whole cross-section. We may anticipate then, that with our convergent nozzle the

\* Dr Tietjens informs me that the calculations, on which the figure given in his book<sup>(3)</sup> is based, were carried out by himself, but that details have never been published.

velocity-distribution across the orifice will be practically square-headed. This anticipation is confirmed when we come to compare the momentum, as deduced from velocity-profile measurements in the jet (where  $J = 1.047 A^2/B$ ) with that calculated from the volume efflux  $Q$  per second at the corresponding pressure-head. To find  $J$  from  $Q$  we must know the velocity-distribution at the orifice; for a parabolic distribution the momentum  $J_P = \frac{4}{3} \rho Q^2 / \pi a^2$ , while for a square-headed distribution the momentum  $J_S = \rho Q^2 / \pi a^2$ . In figure 6 the two curves  $J_P$  and  $J_S$ , computed from the experimental curve of  $Q$  against  $p$ , are shown, and it will be seen that, except from the smallest values of  $R$ , the points deduced from the velocity-profile agree closely with  $J_S$ , indicating a uniform velocity across the jet. It would appear that the parabolic distribution with our particular jet ceases when  $R =$  about 80 or less, and the uniform velocity-distribution is certainly in force when  $R >$  about 180.

### § 5. EFFECTIVE ORIGIN OF THE JET

Referring to equation (3), we see that for a given value of  $x$  the ratio  $A/B$  should be independent of the momentum of the jet. Our experimental results show, however, that  $A/B$  increases as the momentum increases, as is shown in figure 7, where the dimensionless quantity  $(va)^{-1} A/B$  is plotted against  $R$ . It is clear that if the jet is to be considered as coming from a point, as the calculation assumes, then that point must be taken not at the mouth of the jet but within it, and the distance

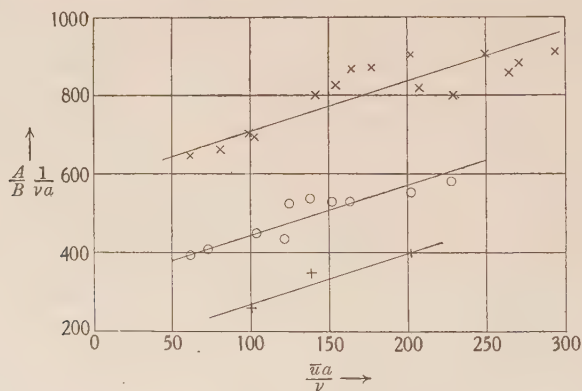


Figure 7.  $\times$ ,  $x = 3.3$  cm.;  $\circ$ ,  $x = 1.8$  cm.;  $+$ ,  $x = 0.8$  cm.

$x_0$  of this imagined point source from the orifice must increase as the momentum increases and the angle of the jet becomes smaller. The simplest assumption, which will be justified by considerations of the kinetic energy, is that  $x_0$  is proportional to  $Q$ , or, what is the same thing, to  $\bar{u}$ , so that

$$A/B = 8v(x + x_0)$$

or

$$\frac{1}{va} \frac{A}{B} = \frac{8}{a}(x + x_0) = 8 \left( \frac{x}{a} + \frac{x_0}{a} \right).$$

This enables us to find a value of  $x_0$  corresponding to each value of  $A/B$ . In figure 8,  $x_0 a$  is plotted against  $\bar{u}a/v$ , and it will be seen that the values obtained at the three



different values of  $x$  are quite consistent. The points show a certain scatter, but it is clear that the method of fixing  $x_0$  makes severe demands on the data, and that discrepancies of a millimetre or two, that is, of 10 to 20 per cent, are to be anticipated. The results can be roughly represented by a straight-line relationship, shown by the full line in the figure, which gives

$$\frac{x_0}{a} = 0.160 \frac{\bar{u}a}{\nu} = 0.160 R \quad \dots\dots(5)$$

or

$$\frac{1}{\nu a} \frac{A}{B} = 8 \left( \frac{x}{a} + 0.160 \frac{\bar{u}a}{\nu} \right).$$

The three straight lines in figure 7 correspond to this formula, and it will be seen that they represent the experimental results as well as can be expected. In particular one value of  $x_0$ , corresponding to a given value of  $\bar{u}a/\nu$ , represents the results at all

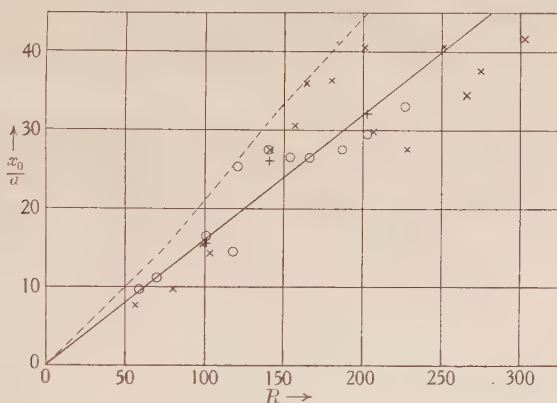


Figure 8.  $\times$ ,  $x=3.3$  cm.;  $\circ$ ,  $x=1.8$  cm.;  $+$ ,  $x=0.8$  cm.

three values of  $x$ : the separation of the three straight lines, which all have the same slope, is not arbitrary, but given by the formula.

In view of the conditions at the orifice, it might at first sight be supposed that the distance of the effective point source within the tube would be such as to make the theoretical lines of flow at the boundary of the orifice continuous with the walls of the tube; that is, that at this boundary  $v=0$  and  $\partial v/\partial x=0$  for the flow from the point. On reference to the expression for  $v$  in equation (1), it is seen that  $v=0$  if  $\xi=2$ , which also gives  $\partial v/\partial x=0$ . From the expression for  $\xi$  we find, remembering that  $\rho=1$  and

$$J_s = \frac{\rho Q^2}{\pi a^2} = \pi v^2 R^2,$$

$$\frac{\sqrt{3}}{4} R \frac{y}{x} = 2 = \frac{\sqrt{3}}{4} R \frac{a}{x_0} \text{ at the orifice}$$

or

$$\frac{x_0}{a} = \frac{\sqrt{3}}{8} R = 0.216 R.$$

This neglects, however, the fact that the tube and its baffle will interrupt the theoretical lines of flow due to the point source, so that the constant multiple of  $R$  cannot

be expected to have the value 0.216. It is of interest to note, however, that these considerations give a formula of the same type as equation (5).

To illustrate this point figure 9 has been prepared. It shows the lines of flow from the point  $P$ , taken at a distance  $x_0$  within the jet given by equation (5), the value of  $R$  being 200. The lines are shown broken where they lie within the jet and baffle. It is clear that in the liquid half space they cannot have exactly the form shown near the baffle, but it is also clear that only a slight modification is required to make them conform to the actual boundary conditions. The continuity of the tube of flow  $FF$  with the walls of the jet appears to the eye to be quite good, but actually the point at which  $v=0$  is some little distance outside the jet, and is marked with an arrow. The diagram makes it easy to see how it is that the flow from a point source suitably

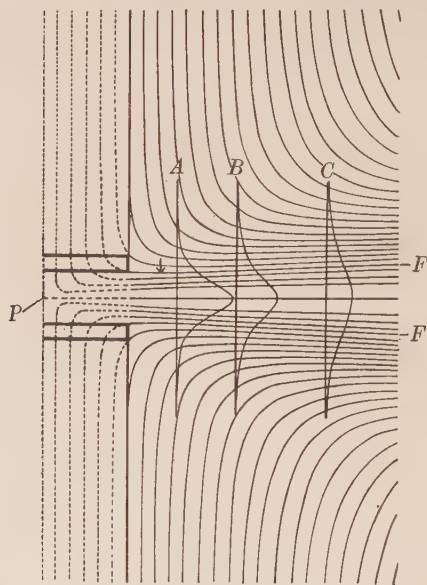


Figure 9.

chosen gives a good representation of the actual flow, except in the immediate neighbourhood of the jet. For completeness the velocity-profiles at the three distances  $A$ ,  $B$  and  $C$  are given. The scale of  $x$  is ten times that of  $y$ .

#### § 6. THE KINETIC ENERGY

If we assume that the velocity at a distance  $x$  from the orifice is given by

$$u = \frac{3}{8\pi} \cdot \frac{J}{\eta x'} \cdot \frac{1}{\left(1 + C \frac{y^2}{x'^2}\right)^2},$$

where

$$C = \frac{3}{64\pi} \cdot \frac{J}{\rho \nu^2} \text{ and } x' = x + x_0,$$

we can derive an expression for the kinetic energy in terms of  $x_0$ . Schlichting's calculation with a point source gives, of course, an infinite velocity and energy at the orifice.

The kinetic energy  $E$  passing per second through a plane distant  $x$  from the end of the tube is given by

$$\begin{aligned} E &= \pi \rho \int_0^\infty u^3 y dy \\ &= \frac{27\rho}{512\pi^2} \frac{J^3}{\eta^3 x'^3} \frac{x'^2}{10C} \\ &= \frac{9}{80\pi} \frac{J^2}{\nu \rho x'} = \frac{9}{80\pi} \frac{J^2}{\nu \rho (x + x_0)} \end{aligned} \quad \dots\dots(6),$$

or at the orifice

$$E_0 = \frac{9}{80\pi} \frac{J^2}{\nu \rho x_0} \quad \dots\dots(7).$$

A value for  $x_0$  may be obtained by comparing this with the kinetic energy at the orifice as determined by the flow in the tube, which will, however, depend upon the velocity profile assumed. For the parabolic distribution at the orifice

$$J_s = \frac{4}{3} \frac{\rho Q^2}{\pi a^2}$$

and

$$E_0 = \frac{\rho Q^3}{\pi^2 a^4} = \frac{9}{16} \frac{J^2}{\rho Q},$$

while for the rectangular distribution

$$J_s = \frac{\rho Q^2}{\pi a^2} \text{ and } E_0' = \frac{1}{2} \frac{\rho Q^3}{\pi^2 a^4} = \frac{1}{2} \frac{J^2}{\rho Q}.$$

Comparing these values with equation (7) we have for the parabolic profile

$$x_0 = \frac{1}{5\pi\nu} Q \text{ or } \frac{x_0}{a} = 0.200 R \quad \dots\dots(8),$$

and for the rectangular profile

$$x_0 = \frac{9}{40\pi\nu} Q \text{ or } \frac{x_0}{a} = 0.225 R \quad \dots\dots(9),$$

so that the difference between the two cases is not large. As we have seen, when  $R < 80$  the value of  $x_0$  should be given by equation (8), while when  $R > 180$ , or earlier, it should be given by equation (9). The broken line, figure 8, represents the values of  $x_0$  which comply with these conditions. At the higher values of  $R$  the values of  $x_0$  derived from the momentum conditions are nearly 30 per cent smaller.

The reason for this discrepancy lies in the nature of the orifice conditions given by the assumption of a point origin within the tube. The actual velocity profile at the orifice varies from parabolic to square, according to  $R$ , while the assumption of a point source within the tube must give at the orifice a profile of the form shown in figures 4 and 5. Figure 10 shows, for a particular value of  $R$ , the profile from a point source compared with the parabolic and square profiles which give, about the  $x$  axis, the same second moment for the enclosed area, i.e. the same momentum at the orifice. The kinetic energy given by the actual profile must clearly, in this case, be less than that given by the assumption of a point source, or, in other words, we must on this assumption increase  $x_0$  to get the right kinetic energy.

The difference between the value of  $x_0$  given by the momentum distribution and by comparison of the kinetic energies at the orifice is, then, due to the fact that the latter method involves the distribution at the orifice itself. When the distance from the orifice exceeds a certain value (which is not greater than 0.8 cm. with our jet of diameter 0.09 cm.) the velocity-distribution, and therefore the kinetic energy, is given to a close approximation by substituting the value of  $x_0$  given by equation (5) in

$$E = \frac{9}{80} \frac{\rho Q^4}{\pi^3 a^4 \nu} \frac{1}{x + x_0}$$

if the distribution at the orifice is rectangular ( $R > 180$  with our type of jet). The energy is only  $\frac{9}{8}$  times greater if the distribution is parabolic.

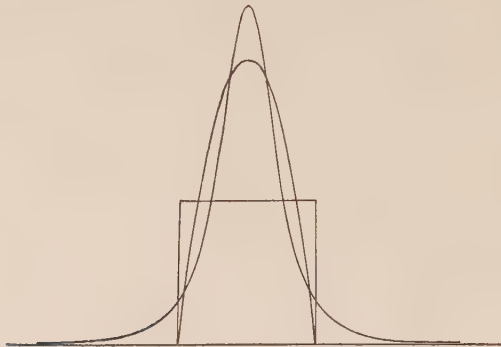


Figure 10.

As regards the loss of kinetic energy which takes place in the jet, for larger values of  $R$  we have then

$$\frac{E}{E_0} = \frac{x_0 + x}{x_0} = 1 + \frac{x}{0.225 Ra}.$$

#### § 7. ACKNOWLEDGEMENTS

We have had the advantage of discussing certain points with Professor L. N. G. Filon, F.R.S., to whom our best thanks are due. The lenses used in the photography were originally purchased from a grant generously made by the Imperial Chemical Industries, Ltd.

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## DISCUSSION

Mr J. H. AWBERY. I wonder whether Prof. Andrade would consider that the formula given for the effective position of the source can be applied generally, even with jets of somewhat different convergence, or whether the coefficient 0.16 is dependent on this factor.

Dr A. B. WOOD. A point of great interest is centred at the orifice of the tube, and it would be valuable if the authors could extend their observations (if they have not already done so) to the liquid flow near this point, just inside and just outside the tube.

Dr J. E. R. CONSTABLE. With regard to the authors' deduction that, except for low rates of flow, there was an approximately uniform velocity-distribution in the water flowing in the tubes they tested, I should like to ask whether they expect a similar distribution in the gas emerging from a sensitive-flame jet. I understand that the work described in this paper had its inception in a study of the action of sensitive flames.

Miss T. J. DILLON. Could this method be applied to experiments on a blast of air from a jet if suitably light particles were used?

S. R. BENNETT. I should like to ask the authors whether the method could be applied to distinguish between the particles originating in the jet itself and those entrained from the surrounding fluid.

AUTHORS' reply. In reply to Mr Awbery and Dr Constable: As long as the velocity is uniform across the orifice the coefficient 0.160 will be appropriate. As is pointed out in the paper, this uniformity of velocity is to be expected so long as the Reynolds's number  $\bar{u}a/\nu$  is not very small and so long as the jet does not terminate in a very long uniform tube—that is, so long as it is of anything like the form used in our experiments. In reply to Dr Wood: We tried to measure the velocity in the immediate neighbourhood of the orifice as has been suggested by Dr Wood, but were not successful. We are not clear as to the reason for the unsatisfactory nature of our plates for this region. In reply to Miss Dillon: The method could undoubtedly be applied to a non-turbulent air blast. In reply to Mr Bennett: In order to differentiate between the particles originating in the jet and those entrained, some colouring matter should be introduced into the liquid which issues from the jet. This then distinguishes it from the liquid through which it flows, but does not obscure the particles if a suitable concentration is chosen. We have actually done this in certain experiments.

## SECONDARY-ELECTRON EMISSION FROM COMPLEX SURFACES

BY L. R. G. TRELOAR, B.Sc., F.INST.P.

(Communication from the Research Staff of the M.O. Valve Company, Limited,  
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**ABSTRACT.** The secondary emission from metals on whose surfaces foreign substances (e.g. electropositive metals) have been deposited to various thicknesses up to a few atomic layers has been studied. It is shown experimentally that for sufficiently thin films of the contaminant the secondary emission for a given base metal depends only on the work function of the surface, and not at all on the nature of the contaminant. The form and magnitude of the variation of secondary emission with work function agree with those calculated from the energy-distribution of secondary electrons. From a study of thicker films information is obtained concerning the mean depth of liberation of secondary electrons. The significance of the results in connexion with more complex secondary-emitting materials is discussed.

### § 1. INTRODUCTION

THE possibility of making use of secondary-electron emission to obtain an amplification of electronic currents has led to a search for materials having high secondary-emission coefficients. Thus it has been found that certain metallic oxides, after treatment with alkali metal vapours, yield secondary-emission coefficients very much higher than any that have been obtained from clean metals. One of the best of these emitters is caesium on caesium oxide on silver, formed by heating oxidized silver at 200° C. in caesium vapour. This material, which is frequently employed as a photoelectric cathode, has been found by Penning and Kruithof<sup>(1)</sup> to give a secondary-emission coefficient of between 8 and 10 at 500 volts, and similar values have been reported by other workers.

It is clear that the materials which give these high secondary-emission values have also very low work functions. It is probable, therefore, that the work function must be regarded as one of the fundamental physical factors by which the magnitude of the secondary-emission coefficient is determined. But it is not necessarily the only factor. It is the purpose of the present investigation to determine to what extent changes of work function, unaccompanied (so far as this is possible) by other changes, affect the secondary-emission coefficient.

### § 2. METHOD OF TREATMENT

The work function of a material may be changed by depositing on its surface a thin film of a different material. Thus, for example, the work function of tungsten may be reduced by several electron-volts by the adsorption on its surface of a

barium layer less than one atom thick, and the thermionic emission is increased by a large factor as a result of the reduction in the energy which an electron is required to possess in order to pass through the surface.

When a metal is bombarded with electrons the secondary electrons, which originate beneath the surface<sup>(2)</sup>, must lose energy in passing through the surface, in exactly the same way as the thermally emitted electrons. It is to be expected that, in general, there will be some secondary electrons approaching the surface from within which have insufficient energy to escape; these electrons will fall back into the interior of the metal. The relative number of such electrons will clearly depend on the work function, becoming smaller as the work function is reduced. We should therefore expect the relative number escaping, i.e. the secondary-emission coefficient, to increase if the work function of the metal is reduced, for example, by the presence of an adsorbed layer of foreign atoms.

Furthermore, given a knowledge of the energy-distribution of the secondary electrons, it should be possible to calculate how the relative number of electrons escaping will be affected by a given change in the work function.

This calculation is made in the following pages, and the resulting relationship between secondary emission and work function is compared with that found experimentally for a metal contaminated with different substances.

### § 3. THEORETICAL EXAMINATION OF THE RELATION BETWEEN SECONDARY EMISSION AND WORK FUNCTION

*The general case.* The argument outlined in the preceding paragraph, which will now be developed quantitatively, depends for its validity upon the assumption that the adsorbed film itself neither absorbs primary electrons nor emits secondary electrons. Any effect which it has on the secondary-emission coefficient will be considered to take place solely through the agency of the accompanying changes in work function.

The energy corresponding to the component of velocity normal to the surface will be referred to as the *normal energy* of the secondary electrons. In considering the effect of the work function on the energy of the escaping secondary electrons it is the normal energy only which will be modified, since there are no electrical forces parallel to the surface.

The normal energy distribution of the secondary electrons escaping from a clean metal may be written in the form

$$dn = f(u) du \quad \dots\dots(1),$$

where  $dn$  is the number of electrons having normal energy between  $u$  and  $u + du$ , measured in electron-volts.

The total number having normal energy greater than any positive value  $U$  will be

$$n = \int_U^{\infty} f(u) du \quad \dots\dots(2).$$

If the work function of the metal is now increased by an amount  $\chi$  eV. by a suitable contamination, only those electrons which formerly escaped from the clean

metal with normal energy greater than  $\chi$  will now be able to escape, and the number  $n$  of such electrons will be given by

$$n = \int_{\chi}^{\infty} f(u) du \quad \dots\dots(3).$$

If  $\delta_0$  and  $\delta$  are the secondary-emission coefficients for the clean and contaminated metal respectively, it follows that

$$\frac{\delta}{\delta_0} = \frac{\int_{\chi}^{\infty} f(u) du}{\int_0^{\infty} f(u) du} \quad \dots\dots(4).$$

It is therefore possible by means of equation (4) to calculate the effect of an increase of work function on the secondary-emission coefficient, provided that the normal energy distribution of the secondary electrons from the clean metal is known.

The effect of a reduction of work function, with which this paper is mainly concerned, cannot be calculated with the same accuracy, since it is necessary to extrapolate the distribution function represented by equation (1) into the region of smaller values of energy, for which  $u$  is negative. (The quantity  $u$  in equation (1) represents the energy of the electrons outside the metal. An electron whose energy within the metal is less than the minimum energy which it must possess in order to escape by an amount  $u$  may therefore be said to possess an energy  $-u$ .) If such an extrapolation can be made, the secondary-emission coefficient of the contaminated surface may likewise be deduced from equation (4), but in this case the change of work function  $\chi$  will be negative.

*Application to Maxwellian distribution.* If the secondary electrons have an energy-distribution represented by Maxwell's Law, equation (1) becomes

$$du = Ae^{-\alpha u} du \quad \dots\dots(5),$$

where  $A$  and  $\alpha$  are constants. Substitution in (4) gives

$$\frac{\delta}{\delta_0} = \frac{A}{\alpha} e^{-\alpha\chi}$$

or

$$\log_{10} \delta = C - b\chi \quad \dots\dots(6),$$

where  $C$  and  $b$  are constants,  $\delta_0$  being fixed. If in this equation  $\chi$  is taken as the independent variable, it is seen that for a given metal, variously contaminated, the logarithm of the secondary-emission coefficient varies linearly with the work function.

An equation equivalent to equation (6) has been derived theoretically by Sixtus<sup>(3)</sup> for the special case of a Maxwellian distribution of energy. Sixtus also measured the secondary-emission coefficient and work function for a thoriated tungsten filament in different states of activation. He found the equation to be applicable over a limited range of work functions, but the coefficient  $b$  was found to vary greatly with the energy of the primary electrons. These results will be considered in greater detail later.



*Derivation of normal energy-distribution.* Before considering the applicability of the results deduced in the preceding sections, it is necessary to examine the available data on the energy-distribution of secondary electrons. The first point to be noted is that there are no experimental data for the *normal* energy-distribution. Such measurements as have been made on the energy-distribution fall into the following two classes.

(a) The retarding potential method. In this method the secondary electrons from a target are collected by a chamber (usually spherical) surrounding it, to which negative potentials with respect to the target are applied. Provided that the chamber is large compared with the target, this method gives the integral of the *total* energy-distribution curve, i.e. the number of electrons having total energy greater than any given value.

(b) The magnetic-spectrum method. In this method a beam of secondary electrons from a target is deviated by a magnetic field, and the total energy-distribution is obtained directly.

The most accurate work on the total energy-distribution is probably that of Haworth, who has obtained curves for molybdenum<sup>(4)</sup> and columbium<sup>(5)</sup> by method (b). His data for molybdenum will be taken as a basis for the calculations which follow. Curve *a* in figure 1 is taken from Haworth's paper, and shows the total energy-distribution curve up to an energy of 35 eV. This portion of the curve was found to be nearly independent of the primary energy for primary energies greater than 37 eV.

In order to be able to derive the normal energy-distribution from the total energy-distribution, it is necessary to make some assumptions about the distribution in angle of the secondary electrons. On this point the experimental evidence is not conclusive, but it suggests that the number of secondaries emitted per unit solid angle in a direction making an angle  $\theta$  with the normal is proportional to  $\cos \theta$ <sup>(6)</sup>. This law will be assumed to be true, and it will be assumed also to apply to any fraction of the emitted electrons having total energies within any given range. The second of these assumptions is equivalent to the statement that the energy-distribution function is the same for beams of electrons emitted in any direction.

If now we suppose the number of electrons emitted, independently of direction, with energy between  $V$  and  $V+dV$  to be given by

$$dN = F(V) dV \quad \dots\dots(7),$$

then, from the cosine law, the number having total energy within this range which are emitted at angles to the normal greater than  $\theta$  may readily be shown to be  $dN \cos^2 \theta$ . Of the electrons having total energy between  $V$  and  $V+dV$  these, and these only, have normal energy less than  $V \cos^2 \theta$ . If we now write  $V \cos^2 \theta = U$ , where  $U$  is a constant, we obtain for the number having total energy between  $V$  and  $V+dV$  and normal energy less than  $U$ , where  $V > U$ ,

$$dN \cos^2 \theta = dN \cdot U/V = U \cdot F(V) dV/V \quad \dots\dots(8),$$

from equation (7).

The total number having normal energy less than  $U$  and any value of total energy greater than  $U$  is found by integrating (8) from  $U$  to  $\infty$ , i.e.

$$U \int_U^{\infty} \frac{F(V) dV}{V} \quad \dots\dots(9)^*$$

And of course all those electrons for which  $V < U$  have normal energy less than  $U$ . Adding this quantity to equation (9), we find that the total number having normal energy less than a given value  $U$  and any value of total energy is

$$\int_0^U F(V) dV + U \int_U^{\infty} \frac{F(V) dV}{V} \quad \dots\dots(10).$$

From this it may readily be deduced that the number  $N_U$  having any value of total energy and normal energy greater than  $U$  is given by

$$N_U = \int_U^{\infty} F(V) dV - U \int_U^{\infty} \frac{F(V) dV}{V} \quad \dots\dots(11).$$

Since  $N_U$  in equation (11) and  $n$  in equation (2) both represent the number of electrons having normal energy greater than  $U$ , the right-hand sides of these two equations must be equal, and the relation between secondary-emission coefficient and work function is given by an equation analogous to equation (4), namely

$$\delta/\delta_0 = N_{\chi}/N_0 \quad \dots\dots(12),$$

where  $\chi$  is the increase of work function produced by a contamination.

Starting from Haworth's curve for the total energy-distribution of secondary electrons for a primary energy of 150 eV., and determining the values of the integrals in equation (11) graphically, we obtain the full curve shown in figure 2 when  $\log_{10} N_U$  is plotted against  $U$ . The portion of the curve shown covers a range of 5 eV., which includes 52 per cent of the secondary electrons.

It will be observed that the curve thus obtained approximates closely to the Maxwellian form given by  $\log_{10} N_U = A - bU$ . The value of  $b$  which best fits the curve over the range shown is found from the slope of the dotted line in figure 2 to be  $0.070 \text{ (eV.)}^{-1}$ . This should therefore also be the value of  $b$  in equation (6), representing the relation between secondary-emission coefficient and work function, in the case of contaminated molybdenum. Figure 1 (b) shows a Maxwellian distribution of total energy corresponding to this same value of  $b$ . It is noteworthy that the integral of the normal energy-distribution curve approaches the Maxwellian form more nearly than does the total energy-distribution.

*Deductions from the foregoing theory.* The experiments to be described were carried out with the object of testing the applicability of the foregoing theory to the case of a contaminated metal surface. The following four main results are to be expected: (1) A linear relationship should exist between  $\log_{10} \delta$  and  $\chi$ . (2) The constant  $b$ , representing the rate of change of  $\log_{10} \delta$  with  $\chi$ , should have the value  $0.070 \text{ (eV.)}^{-1}$  in the case of molybdenum. (3) For a given metal, the value of  $b$  should be independent of the nature of the contaminant. (4) For a given metal, the value of  $b$  should be nearly independent of the energy of the primary electrons.

## § 4. EXPERIMENTAL METHOD

In carrying out the experiments, use was made of the method of measuring secondary emission from filaments described by the author in an earlier paper<sup>(7)</sup>. A valuable feature of the method is that it permits of the measurement of changes in the relative contact potential of the filament whose secondary emission is being studied, with respect to a clean tungsten filament. The changes of contact potential give a measure of the changes in work function.

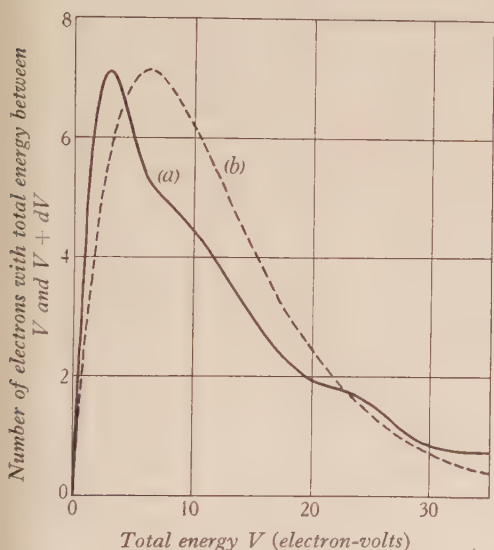


Figure 1. Total energy-distribution curves: (a) Secondary electrons (Haworth), (b) Maxwellian.

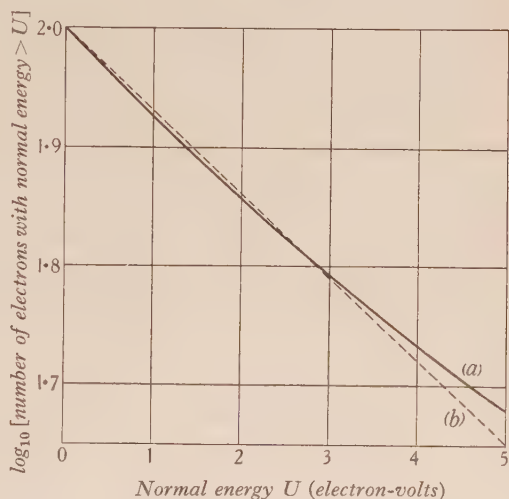


Figure 2. Integrals of normal energy-distribution curves: (a) Secondary electrons, (b) Maxwellian.

The cross-section of a typical tube is shown in figure 3 (a), the main dimensions being the same as those given in the paper previously referred to. The tungsten cathode *C* provides the primary current, *T* is the target (secondary-emitting) filament, and *B* an extra filament to provide the contamination, e.g. an oxide-coated nickel filament. This filament was maintained at cathode potential during the secondary-emission measurements.

*The contact-potential measurements.* The measurement of the contact potential was carried out in the following way. The cathode was maintained at a temperature such that a saturated thermionic emission current of 5 mA. was drawn to the anode and grid, when these two electrodes were at 100 volts positive potential with respect to the cathode. The target current was then measured, as a function of the target potential, with respect to the cathode, over a range of a few volts above that at which it first assumed a measurable value. The curve obtained under these conditions had a considerable linear portion, which, when extrapolated, cut the zero current axis at a point *x*. The difference between the value of *x* for the uncontaminated target and its value for any state of contamination was taken to be equal

to the contact potential-difference between the clean and contaminated surfaces. It is also equal to the difference between their work functions.

It was found that the value of  $x$  obtained for any state of the target was not affected by changes of a few volts in the potential applied to the anode and grid. Any changes occurring in the contact potential-difference between these electrodes and the cathode could therefore be neglected.

The value of  $x$  obtained in this manner was referred to the potential of the tungsten cathode. In order that changes in  $x$  should give a true measure of the changes in contact potential of the target, it was necessary to ensure that the cathode was always in the same condition when the measurement was carried out. Since the cathode, as well as the target, became contaminated during the experiments, it was therefore necessary to remove this contamination by heating the cathode to a high temperature before taking each set of readings.

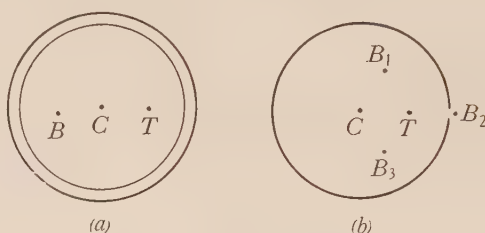


Figure 3. Cross-sections of electrode systems.

## § 5. EXPERIMENTAL RESULTS

The experimental results for films of barium on molybdenum, and of barium, thorium and oxygen on tungsten will now be presented, and a comparison of the results with the theoretical expectations will follow. An examination of some of the effects produced by comparatively thick films will then be made.

*Barium (from an oxide-coated filament) on tungsten.* Figure 3 (a) shows the disposition of the electrodes in this experiment.  $T$  was a tungsten filament 0.104 mm. in diameter and 50 mm. long,  $C$  was a similar filament 12 mm. long, and  $B$  was a nickel wire 0.18 mm. in diameter and 50 mm. long, coated with a mixture of barium and strontium carbonates in equal molecular proportions. During the pumping of the tube the filament  $B$  was heated to convert the carbonates to oxides, and eddy-current heating and barium gettering were carried out before sealing off the tube. (A slight modification of the original design, introduced during the course of the work, consisted in making the grid free to slide at one end instead of rigidly fixed. This precaution was taken to prevent distortion of the grid during the eddy-current heating.)

In carrying out the experiment, the target and cathode were first cleaned by heating at 2800° K. for a few minutes, and the contact potential and secondary-emission coefficient for 300 volts bombarding potential were measured. The result



of these measurements is indicated by the point *A* in curve *a*, figure 4, which shows the relation between  $\log_{10} \delta$  and the change in the work function of the target. The oxide-coated filament was then heated at about 1400° K. for a short time, and after the cathode had been cleaned the measurements were repeated. This second point is shown at *B*. The deposition of barium on the target from the oxide-coated filament had caused a reduction in the work function and an increase in the secondary emission. Successive repetition of these operations gave the points indicated by crosses, the final point being *C*.

On now heating the target at successively higher temperatures so as to re-evaporate the barium, the points marked by circles were obtained. The final condition of the target (point *D*) was nearly the same as its initial condition.

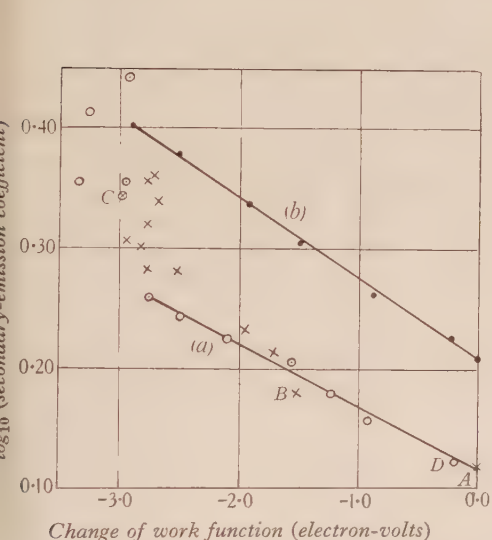


Figure 4. Relation between secondary-emission coefficient and work function: (a) Barium (from oxide-coated filament) on tungsten,  $\times$  deposition,  $\circ$  evaporation. (b) Barium on molybdenum. Primary-electron voltage 300.

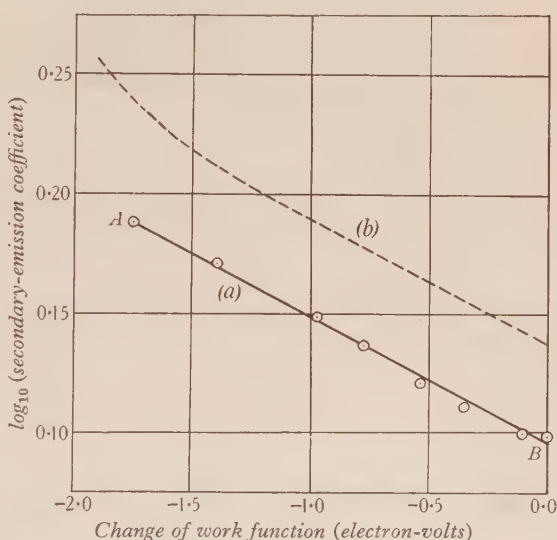


Figure 5. Relation between secondary-emission coefficient and work function for thoriated tungsten: (a) Author's results, (b) Sixtus's results. Primary-electron voltage 300.

*Barium (from an oxide-coated filament) on molybdenum.* This experiment was in all respects similar to that described for tungsten, except that points were obtained only during the later stages of evaporation of barium from the target. The results of this experiment, which is shown in curve *b*, figure 4, have already been published in *Nature*<sup>(8)</sup>.

*Thorium on tungsten.* In this experiment the target was a thoriated tungsten filament of diameter 0.1 mm. The contaminant, thorium, was brought to the surface of the filament by diffusion from within the metal. In curve *a*, figure 5, the point *A* represents the fully activated filament. The remaining points were obtained during deactivation at successively higher temperatures. The final point *B* represents a clean (or very nearly clean) surface of tungsten, and it is noteworthy that the

value of  $\delta$  for this condition is very nearly the same as that found for pure tungsten, point *A*, figure 4 (*a*). A similar experiment has been performed by Sixtus<sup>(3)</sup>, and it is interesting to compare his results, which are reproduced in curve *b*, figure 5, with the author's.\*

*Oxygen on tungsten.* The tube used in this experiment had attached to it a side tube containing a small quantity of potassium permanganate. After measurement of the secondary-emission coefficient at a primary voltage of 300, and of the contact potential of the target, which was of tungsten, the temperature of the target was raised to 1500° K. and oxygen was admitted by heating the permanganate. After the gas had been absorbed by the getter, measurements were made on the oxygenated filament.

The following results were obtained:

For clean tungsten  $\delta$  = 1.31.

For oxygenated tungsten  $\delta$  = 1.06.

Change of work function on oxygenation = 1.78 eV.

The value for the contact potential difference between clean and oxygenated tungsten is in close agreement with Reimann's value of 1.75 V.<sup>(9)</sup>

#### § 6. DEDUCTIONS FROM THE EXPERIMENTS

The results for barium on tungsten, barium on molybdenum, and thorium on tungsten prove that, provided that the contaminating film is not too thick, there exists a linear relationship between  $\log_{10} \delta$  and  $\chi$ , in accordance with the theoretical expectation. The region representing thicker films in figure 4 (*a*) will be considered later.

The slopes of the curves, giving the values of *b* in equation (6), are given in table 1 for the primary voltages indicated.

Table 1. Values (eV.<sup>-1</sup>) of *b* for various contaminated surfaces

Voltage (V.)	300	150	35
Barium on molybdenum	·067	·057	·050
Barium on tungsten	·052	·060	·043
Thorium on tungsten	·053	—	—
Oxygen on tungsten	·052	—	—

From these figures it is clear that for a given value of the work function and for the different contaminations on tungsten, the secondary emission is independent of the nature of the contamination. The value of *b* is found to change only to a relatively slight extent with variations in the primary voltage. The variations in table 1 are hardly outside the experimental errors.

These variations do not accord with the data given by Sixtus for thoriated tungsten<sup>(3)</sup>. He found the value of *b* to fall considerably as the primary voltage was

\* The author regrets that at the time of reporting his first results to *Nature* he was not aware of the work of Sixtus.

increased from 200 to 800. Thus at 400 V., for example, its value was 40 per cent lower than at 200 V. No such large variation is to be expected from the theoretical considerations brought forward earlier in this paper.

#### § 7. COMPARISON OF THEORETICAL AND EXPERIMENTAL VALUES OF $b$

It has been shown that the experimental results are concordant with the main predictions of the theory. The quantitative comparison between the theoretical and experimental values of  $b$  remains to be made.

The theoretical value was  $0.070 \text{ (eV.)}^{-1}$  for molybdenum at 150 eV. primary energy.

An examination of the experimental data gives reason to believe that the difference between the 150-V. and 300-V. values for molybdenum shown in table 1 is fortuitous. The mean of these two values will therefore be taken as the most probable experimental value of  $b$  for 150-V. primaries. The value thus obtained is  $0.062 \text{ (eV.)}^{-1}$ , which is about 12 per cent less than the theoretical value.

It will be remembered that the theoretical value was arrived at from the slope of the line which best fitted the integrated normal-energy-distribution curve over an arbitrary range of positive energies. The experiments are concerned with the region of negative energies (as defined in § 3) covering the approximate range 0 to  $-3 \text{ eV.}$  A more exact comparison of the theoretical and experimental values therefore necessitates finding the mean slope of the extrapolated curve of figure 2 over the range 0 to  $-3 \text{ eV.}$  This extrapolation, though it cannot be made very exactly, yields the value  $0.085 \text{ (eV.)}^{-1}$  for  $b$ , a value which should be nearer the truth than  $0.070$ . With this revised value of  $b$ , the experimental value is 27 per cent low.

The experimental errors involved in the determination of  $b$  are rather large. The theoretical value involves the errors in the measurement of the energy-distribution and the error introduced by the extrapolation to negative energies. Even when all these errors are considered, however, it is difficult to account for a difference of 27 per cent.

In considering the possible causes of this discrepancy, it must be remembered that the validity of the assumption of a cosine law of distribution in angle of the secondary electrons cannot be regarded as established beyond doubt.

#### § 8. EFFECTS WITH THICK FILMS

Our interest thus far has been confined to the thin-film contaminations for which the linear relationship between  $\log_{10} \delta$  and  $\chi$  is applicable. A special experiment was carried out with the object of investigating the thick-film region. The oxide-coated filament, previously used as a source of barium, was replaced by a nickel-barium alloy wire containing about 0.6 per cent of barium. It was hoped that the barium obtained from this source would be purer than that from an oxide-coated filament,

since it is known that an oxide-coated filament gives off oxygen as well as barium, the relative amount of oxygen depending upon the amount of ageing to which the filament has been subjected, as well as on its temperature<sup>(10)</sup>.

In order to secure a fairly uniform deposition of barium over the surface of the target, three nickel-barium wires  $B_1$ ,  $B_2$  and  $B_3$ , figure 3 (b), were arranged symmetrically round the target  $T$ . To avoid undue complication the grid was omitted, the primary current calibration (required for the measurement of  $\delta$ ) being taken from the dimensionally similar tubes already described, in which the grid was present.

The relation between  $\log_{10} \delta$ , at a primary voltage of 300, and  $\chi$  for this tube is shown in figure 6, which is drawn in a manner comparable with figure 4 (a). The portion  $AB$  of this curve represents the thin-film region, whilst  $BC$  represents the

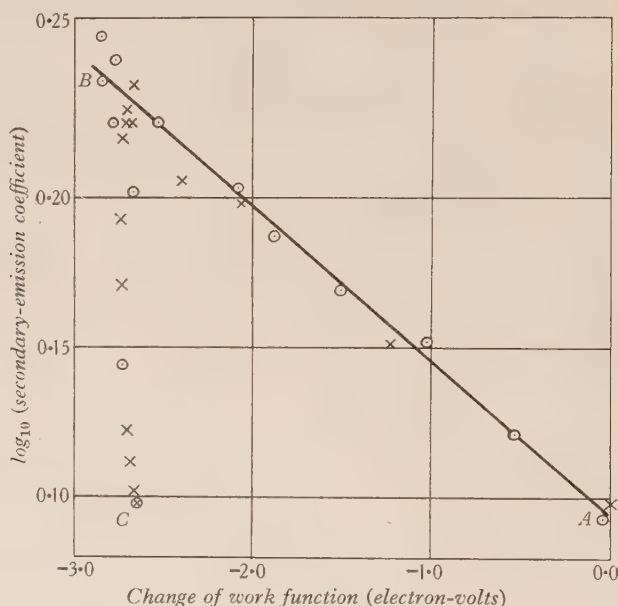


Figure 6. Relation between secondary-emission coefficient and work function. Barium (from nickel-barium alloy) on tungsten:  $\times$  deposition,  $\odot$  evaporation. Primary-electron voltage 300.

thick-film region. The thin-film portion of the curve falls along a straight line whose slope is  $0.052 \text{ (eV.)}^{-1}$ . It is interesting to observe that whereas the thin-film portions of the curves of figure 4 (a) and figure 6 have exactly the same slope, the thick-film portions are entirely different. The thick-film points lie above the linear portion of the curve for barium derived from an oxide-coated filament, and below for barium derived from a nickel-barium alloy. This difference must be attributed to the presence of oxygen, as well as barium, in the former case. This also accounts for the greater maximum reduction of work function produced when the source of the contamination is an oxide-coated filament. It may be mentioned that these differences are quite typical and have been observed with a number of different tubes.



In deriving the theoretical expression for the effect of variations of work function on secondary emission, it was assumed that the contaminating film neither absorbed primary electrons nor emitted secondary electrons. It would appear that the region *BC* in figure 6 represents thicknesses of film for which this assumption is no longer true, and that for points in this region the secondary electrons originate at least partly in the barium film itself.

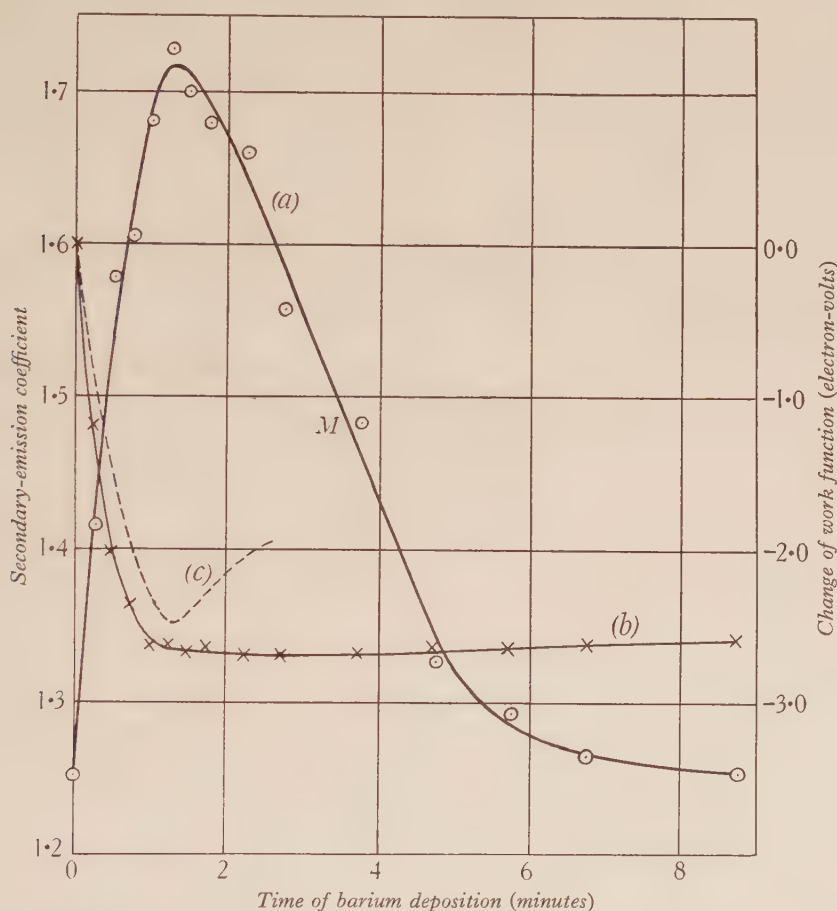


Figure 7. Variation (a) of secondary-emission coefficient and (b) of work function, with amount of barium on surface of tungsten. (c) Becker's work-function curve.

The data from which figure 6 was obtained are represented in a different manner by curve *a*, figure 7. This figure shows the variation of the secondary-emission coefficient with the total time of deposition of barium from the nickel-barium wires. An independent experiment showed that the rate of evaporation of barium was practically constant for the temperature and time used. Hence the time of deposition gives a measure of the relative amount of barium present on the target.

The rising portion of this curve represents the thin-film region, in which the secondary electrons originate almost entirely in the tungsten. The final portion

shows a tendency for the secondary emission to approach a constant value and indicates that here the effect of the underlying tungsten is becoming negligible. In the intermediate (falling) portion of the curve the tungsten and the barium both contribute to the secondary emission.

#### § 9. THE DEPTH AT WHICH SECONDARY ELECTRONS ORIGINATE

At the point *M*, figure 7, where the secondary emission is half-way between the thin-film and thick-film values (the work function being approximately constant) the secondaries probably originate in about equal numbers from the tungsten and barium films. The thickness of film corresponding to this point may therefore be taken to be approximately equal to the mean depth at which the secondary electrons originate.

An attempt will now be made to compute this depth. This may be found in terms of the thickness of film which gives the maximum thermionic emission or minimum work function (subsequently referred to as the *optimum thermionic layer*) by reference to the change in work function for different times of barium deposition, curve *b*. This curve may be compared with Becker's<sup>(11)</sup>, which shows how the reduction of work function varies with the relative surface concentration of barium on tungsten. The absence of a clearly defined minimum in the author's work-function curve is probably associated with non-uniformity of the deposit, as a result of which different portions of the target attained the minimum work function at different times. In Becker's experiments such non-uniformities were probably reduced by diffusion of barium over the surface of the tungsten when the filament was heated. However, by considering the rate of fall of work function in the early stages of the deposition, it is possible to obtain a fairly accurate indication of the time at which the optimum thermionic thickness was effectively reached. This time comes out at 1.3 min. The point *M* therefore corresponds to 2.8 optimum thermionic layers.

If we can now translate depths in terms of optimum thermionic layers into depths in terms of atomic layers, we shall be able to arrive at the mean depth at which the secondary electrons originate. Unfortunately the thickness of film, or surface concentration of barium atoms corresponding to one thermionic layer, has not been experimentally determined. For caesium on tungsten the optimum thermionic layer has been found by Langmuir<sup>(12)</sup> to be 0.67 of a monatomic layer, but there is no reason to believe that the same value will apply for barium on tungsten. From theoretical considerations de Boer<sup>(13)</sup> has estimated the optimum covering for barium to be 0.29 of a monatomic layer. Perhaps the best that can be done is to take a value of 0.5 for this quantity. The mean depth at which the secondary electrons originate, according to this assumption, is 1.4 atomic layers.

A correction of this value is necessitated by the fact that the primary electrons strike the target at various angles of incidence. The mean angle of incidence, under the conditions of measurement (i.e. with target potential 300 and anode potential 450), may be taken to be the same as if the electrons remain undeviated from their rectilinear paths by the presence of the target; its value is therefore about 30° to the

normal. On the basis of Bruining's work<sup>(2)</sup> on the effect of angle of incidence on the mean depth at which secondary electrons are liberated, the depth for normal incidence will thus be increased by the factor  $\sec. 30^\circ$  compared with the depth for  $30^\circ$  incidence. The application of this correction to the value obtained above gives 1.6 atomic layers for the mean depth at which the secondary electrons originate, with a primary voltage of 300 and normal incidence. This figure may be in error by a factor of about 2, on account of the uncertain value of the optimum thermionic layer.

From his experiments Bruining<sup>(2)</sup> has calculated the product of the absorption coefficient for secondary electrons in nickel and the mean depth at which they originate to be 0.45 for 300-V. primary electrons. He then substitutes Becker's value<sup>(14)</sup> of  $1.5 \times 10^6$  for the absorption coefficient for electrons of energy equal to the mean energy of the emitted secondaries, and obtains the value 30 angstroms for the mean depth at which the secondary electrons originate. This figure, which represents about 14 atom-layers of nickel, is of a different order of magnitude from that found in the present work.

#### § 10. OTHER WORK ON COMPLEX SURFACES

It has been shown in § 6 that the effect of a change of work function on the logarithm of the secondary-emission coefficient is nearly independent of the energy of the primary electrons. On this basis it is possible to describe qualitatively how the shape of the curve representing the logarithm of the secondary-emission coefficient as a function of the primary energy will be affected by the deposition on a metal surface of increasing quantities of a contaminant.

So long as the contamination satisfies the thin-film criterion the change of secondary emission will be determined solely by the change in work function, hence the change in  $\log \delta$  will be the same for all values of the primary energy. In this region, therefore, the successive degrees of contamination will be manifested by a displacement of the original curve parallel to the axis of  $\log \delta$ , without alteration of shape. When the thickness of the film becomes sufficient for the effect of the underlying metal to be neglected, a curve will be obtained representing simply the secondary emission of the contaminant in bulk. In the intermediate region, between the thin and very thick films, the shape of the curve will, in general, differ from that for either of the simple materials.

These expectations are substantially verified by the experimental curves of Copeland<sup>(15)</sup> showing the logarithm of the secondary-emission coefficient, as a function of the primary voltage, for a metal on which were deposited successive amounts of a contaminant. In each of the three cases studied, the first effect of the contamination was to displace the curve parallel to the axis of  $\log \delta$ . Thus, in the case of aluminium contaminated with platinum a reduction of  $\log_{10} \delta$  by about 0.15 took place before the shape of the curve became definitely changed. Since the work function of platinum is higher than that of aluminium, a reduction in  $\log \delta$  would be expected as a result of the deposition of the former metal on the latter. An opposite

effect was observed when calcium was deposited on gold; this again is to be expected from consideration of the work function.

Since the thickness of film for which the simple work-function effect ceases to apply is related to the depth at which the secondary electrons are liberated, and since this depth increases with the energy of the primary electrons, it would be expected that the shape of the curve typical of the underlying metal would persist to a greater thickness of film at the higher than at the lower primary energies. This expectation also is borne out by Copeland's results.

Whilst Copeland's interpretation of his experiments is concerned chiefly with what has been described in the present work as the "thick-film" region, it seems that his generalization to the effect that the major portion of the observed changes in secondary emission must be ascribed to some other cause than the variation of work function tends rather to underestimate the importance of a factor which has considerable significance in any interpretation of phenomena of this kind.

#### § 11. THE SIGNIFICANCE OF THE WORK-FUNCTION EFFECT

The general result of the investigation which has been described may be stated in the following terms: *For any metal whose work function is varied by the deposition on its surface of a thin film of a foreign substance, changes in secondary-emission coefficient are determined solely by the accompanying changes in work function. The magnitude of such changes can be predicted if the energy-distribution of the secondary electrons is known.*

There is no reason to doubt that this law can be generalized to include non-metals as well as metals. On the other hand, it does not account for the changes in secondary emission produced by a thick-film contamination, neither does it give any information about the relative secondary-emission coefficients of different specific materials.

The following discussion, which is admittedly speculative, suggests how the knowledge already gained may be brought to bear on the problems raised by some of the highly complex secondary-emitting materials referred to in § 1.

We may begin by considering what is the maximum secondary emission obtainable from a simply-contaminated metal. The secondary-emission coefficient which would be given by tungsten, for 300-V. primary electrons, if its work function were imagined to be reduced to zero by a thin-film contamination would be 2.3, and that for molybdenum under the same conditions would be 2.9. From the knowledge that the energy-distribution curves for secondary electrons for the dozen or so metals that have been studied in this respect are all quantitatively similar, combined with the fact that the range in the secondary-emission coefficients of the clean metals which have been reliably examined is not very great, it is possible to predict that the highest secondary-emission coefficient obtainable from any metal having a thin-film contamination will not be much more than 3.0 at 300 volts, however much its work function is reduced.



It is known that a number of complex substances yield secondary-emission coefficients much in excess of this value. As a typical case caesium on caesium oxide on silver, referred to in § 1, may be cited. The secondary-emission coefficient for this material at 300 V. may be as high as 8.0. There is nothing in the preceding work which gives any hint of the reason for such a high value.

If, however, we regard the material as a thin-film contamination of caesium on caesium oxide, and if we assume that the uncontaminated caesium oxide would have a very much higher work function than the activated surface, there are two interesting possibilities. It may be (a) that the secondary-emission coefficient for the uncontaminated caesium oxide is about the same as that for a metal having the same work function, but the rate of change of secondary-emission coefficient with work function is much greater; or (b) that the rate of change of secondary-emission coefficient with work function is about the same as for a metal, but the secondary-emission coefficient for the uncontaminated caesium oxide is very much greater than that for a metal having the same work function. (b) is unlikely to be true, since no oxides in the uncontaminated state have been reported as having very much higher secondary-emission coefficients than the pure metals. If, however, (a) is true, it follows that the average energy of the secondary electrons from caesium oxide is very much lower than that of the secondary electrons from a metal. This is a matter which could be investigated experimentally.

#### § 12. ACKNOWLEDGEMENT

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## DISCUSSION

Dr J. E. R. CONSTABLE. I should like to ask whether the magnitude of the work function appreciably affects the number of incident electrons which penetrate the target surface and, if so, whether the author has allowed for this effect.

AUTHOR'S reply. If potentials are referred to the cathode, a change in the work function of the target will alter the effective potential outside the metal (i.e. in the vacuum) but not that inside. Owing to the former effect the primary current may be changed by a maximum amount of 0.5 per cent at a primary energy of 300 V., but this has been neglected. Since the potentials within the metal are unaffected, there will be no change in the energy of the primary electrons which penetrate the surface of the target.

# THERMIONIC APPROXIMATIONS TO THE GAS-COVERED FRACTION OF AN ADSORBING SURFACE, APPLIED TO THE TEMPERATURE DEPENDENCE OF OXYGENATION AND OXIDATION OF A TUNGSTEN FILAMENT

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**ABSTRACT.** A distinction is made between two methods of using thermionic data to estimate for a filament surface the fraction which is covered with oxygen. Conditions are formulated under which either of the two gives approximations to the true fraction defined by concentration of particles in an adsorbed layer. In the method depending upon work functions the necessity for obtaining true instead of apparent thermionic constants for a composite surface is introduced, as these are found to differ widely for oxygenated tungsten. A set of experimental data is analysed by means of the two methods in turn, and empirical graphs are thence obtained representing approximately the temperature variation of the gas content of the surface. The limiting slopes to which these graphs tend in the valid portions of their respective ranges agree with the temperature coefficients of oxidation and oxygen evaporation known from other experiments.

## § 1. INTRODUCTION

IN the physics and chemistry of composite surface layers, the fundamental variable is the fraction of surface which is occupied by any given component or which conforms to any given condition. This fraction, written  $\theta$  in most of the literature, has been estimated in restricted cases only, notably the condensation of alkali vapours upon a refractory metal such as tungsten to which they are electropositive and whose thermionic emission they greatly increase. To determine  $\theta$  thermionically for the case of an electronegative gas such as oxygen involves considerably greater difficulty, and has mainly been attempted through its intermediate action in cementing more electropositive materials to the underlying metal. In the course of experiments on reactions at a tungsten surface in oxygen at pressures below  $10^{-3}$  mm. of mercury, it was found necessary to reconsider the relation of  $\theta$  to observable quantities. The results, with their application to the temperature variation of adsorption and oxidation, are presented here. It should be understood that the data are simply saturated thermionic currents obtained with appropriate

precautions in the presence of controllable traces of oxygen too small to involve ionization by collision. In addition to the usual pretreatment of all metal surfaces, high-frequency degassing, baking of walls, etc., and the isolation of an isothermal length of filament by means of guard rings, these precautions included technique for maintaining constancy of very small gas pressures over long periods; details may be found in some already published papers<sup>(1)</sup> on certain other problems for which the experiments were designed.

## § 2. EXPERIMENTAL METHODS FOR OBTAINING AN APPROXIMATION TO THE FRACTIONS OF SURFACE COVERED

In problems in which  $\theta$  is to be a variable in thermodynamic or chemical equations, its appropriate definition is

$$\theta = N/N_1,$$

where  $N$  is the number of gas particles of any one kind per unit area on the surface and  $N_1$  is that number at saturation of the layer. With this definition a plot of  $\log_e \{\theta/(1-\theta)\}$  against  $1/T$  would express by its slope an energy of adsorption or of surface reaction, in the manner familiar in studies of three-dimensional gas behaviour.

But since no means exists for counting the particles which remain on the surface and so correlating  $N$  directly with the accompanying physical changes, an approach must be made to  $\theta$  through some experimental quantity  $\theta_x$  to be chosen as follows. (a)  $\theta_x$  must be unambiguously accessible through some observable property of the material. (b) Whatever the significance chosen for  $x$ , the limits  $\theta_x = 0$ ,  $\theta_x = 1$  must denote respectively the state of bare metal and of complete gas covering, satisfactorily related to some chemical or physical criterion of saturation. (c) The temperature dependence of  $\theta_x$  must follow as closely as possible that of  $N/N_1$ . For thermionic approaches to  $\theta$  the chosen indicator must be the simplest function of emission current  $i$  consistent with the above conditions, so that

$$\theta_x = f(i)$$

and

$$0 < \theta_x < 1.$$

The requirement (c) essentially precludes any one form of  $\theta_x$  from being used over more than a portion of the range  $\theta = 0$  to  $\theta = 1$ , since any simple function which follows the course of  $N/N_1$  when the adsorbed particles are widely separated will fail to do so when they are so closely packed as to influence each other. Hence any variation of  $\log_e \{\theta_x/(1-\theta_x)\}$  which is linear for sparse surface concentrations will become curved before the layer approaches saturation. We proceed to consider two such forms of  $\theta_x$ , or model approximations to  $\theta$ .

(A). If the lateral components of the fields of force surrounding adsorbed particles diminish so rapidly with distance that the effects of any local deposit may be considered to cease abruptly at its edge, the whole surface may be regarded as divided into elementary areas of two kinds, whose electron-emitting properties are those of completely bare and completely gas-covered metal respectively. Using the



Richardson equation, with emission constants  $A_1$  for bare and  $A_2$  for covered surface, and work functions  $\phi_1$  for bare and  $\phi_2$  for covered surface, we have

$$i_\theta = A_1 T^2 e^{-\phi_1/\kappa T} (1 - \theta_x) + A_2 T^2 e^{-\phi_2/\kappa T} (\theta_x) \quad \dots\dots(1).$$

If, for comparable values of  $\theta_x$  and  $1 - \theta_x$ , the first term is large compared with the second term, we have approximately

$$i_\theta = i_0 (1 - \theta_x) \quad \dots\dots(2),$$

whence  $\theta_x$  can be found directly. We adopt the notation  $\theta_a$  for this particular instance of a  $\theta_x$ , determined by current deficiency so that

$$\theta_a = i_0 - i_\theta/i_0 \quad \dots\dots(3).$$

For the highly electronegative deposits with which we deal, the condition  $i_0 \gg i_1$  is fulfilled, oxygen reducing the emission from tungsten to about  $10^{-4}$  of its clean value; for a layer of an electropositive element such as caesium, which greatly increases instead of decreasing the emission, a corresponding  $\theta_x$  would be based on the approximation

$$i_\theta = i_1 \theta_x.$$

(B). If the adsorbed particles modify the field over the whole surface while the layer is still very unsaturated, there is strictly no region behaving as a clean surface, and the whole area may be regarded as altering its properties homogeneously according to the growth of mean surface density of adsorbed material. On this view  $\theta_x$  is not so much a fraction of area as a measure of partial acquisition of a certain property ascribable to the surface as a whole. The equations of paragraph (A) could not then be used unless an inconveniently large number of terms were introduced, with values of  $A$  and  $\phi$  intermediate between those for clean and covered metal.

This second instance of a  $\theta_x$  accordingly appears as a fractional difference in the work done in emission over the whole surface, and we adopt the notation  $\theta_w$ , so that

$$\theta_w = (\phi_\theta - \phi_0)/(\phi_1 - \phi_0) \quad \dots\dots(4).$$

The form in which this definition has been used by Langmuir and others may be seen by substituting in the Richardson equation at constant temperature, obtaining

$$\theta_w = (\log i_\theta - \log i_0)/(\log i_1 - \log i_0) \quad \dots\dots(5).$$

Equation (5) is valid only if  $A$  is constant or if  $\log A$  is proportional to  $\phi$ .

$\theta_w$  has been used in studying the deposition of alkali vapours, where  $i_1$  is large and easily determined. The extremely minute and uncertain values of  $i_1$  for electro-negative layers has hitherto prevented its application to these. This difficulty is escaped only when (as in one of our previous papers)  $i_1$  can be eliminated by isothermal observation at two different values of  $\theta$ .

In general  $\theta_w$  will more nearly approximate to the true  $\theta$ , or  $N/N_1$ , for widely distributed or laterally mobile layers, since they conform more nearly to the physical requirements of paragraph (B), whereas  $\theta_a$  will follow  $\theta$  for immobile material sparsely distributed. Adsorbed vapours consisting of strongly dipolar molecules or inert ions will possess the long-range fields assumed for  $\theta_w$ , while the short-range

fields surrounding neutral atoms will allow the assumptions for  $\theta_a$  to be more nearly correct. These considerations make it clear that neither  $\theta_w$  nor  $\theta_a$  bear the same relation to  $N/N_1$  for large as for small values of  $\theta$ . In particular the contribution made by each additional adsorbed particle to modifying the electronic work function has been shown by several workers to vary with mutual depolarization of neighbours in the surface layer; any such interaction with neighbours will also cause the work of evaporating a neutral particle from the surface to vary with packing of the adsorbed material.

### § 3. $\theta_w$ FOR TRUE WORK FUNCTION AND APPARENT WORK FUNCTION

The sensitiveness of  $\theta_w$  to the value assumed for  $i_1$  in equation (5) shows that this method of approximating to  $\theta$  requires a reliable estimate of the emission constants of the fully covered metal. For the particular interface with which our experiments deal the term *OW* has been used by most writers, without prejudice to the chemical state of the oxygen covering so long as the latter is regarded as complete, so that  $\theta = 1$ . Kingdon<sup>(2)</sup> attempted to determine these constants for *OW* by experiments in which gas was admitted to the hot tungsten filament at a rate sufficient to compensate for oxidation, and his values  $A_{OW} = 5 \times 10^{11}$  A./cm<sup>2</sup> and  $\phi_{OW} = 9.2$  eV. have been widely quoted. Subsequent work, however, has thrown serious doubt on the possibility either of attaining the condition  $\theta = 1$  at any high temperature in the presence of gas, or even of keeping  $\theta$  at any constant value over a temperature range, during rapid oxidation, while Fowler<sup>(3)</sup> and others have shown the inherent improbability of Kingdon's value of  $A_{OW}$  as a genuine thermionic constant. It is generally agreed that  $A_{OW}$  is more likely to be equal to or even less than the  $A_W$  measured with clean metal, about  $10^2$  A./cm<sup>2</sup>; the form of Richardson's equation then shows that observed values of emission demand a much lower value of  $\phi_{OW}$  than that offered by Kingdon. A more reliable way of estimating  $\phi_{OW}$  is due to Reimann's careful and repeatable measurements of the contact potential-difference between *OW* and *W*, since a contact potential represents, to a first approximation, the difference between the values of  $\phi$  for adjacent surfaces.  $\phi_{OW}$  is thus obtainable by adding Reimann's potential to the  $\phi$  for clean tungsten, the result being 6.35 eV. Reimann has shown<sup>(4)</sup> that a quantity of this order can account satisfactorily for the actual small emissions if  $A$  is the same for *OW* as for *W*. Since the contact measurements do not involve the above-mentioned temperature control of  $\theta$  they are free from the objection that applies to Kingdon's experiments, and can replace the latter as the source of  $\phi_{OW}$ .

We have, in the last of the publications referred to, investigated the new interpretation of currents from partially oxygenated tungsten necessitated by the above considerations, and we summarize as follows the portions of our argument relevant to the present need for deducing  $\theta_w$ .

Consider the Richardson plots typified in figure 1, in which the slope of  $\log i/T^2$  against  $1/T$  determines  $\phi$ , while the vertical intercept at zero abscissa is measured to obtain  $\log A$ . The line *bc* for a fully oxygenated surface would, if obtainable,

lie below the line  $bd$  for clean tungsten, and the vertical intercept of  $bc$  would be either identical with or less than that of  $bd$  since the adsorbed oxygen cannot increase emission. But if a trace of oxygen, even below  $10^{-6}$  mm., is present while the readings for the Richardson plot are being obtained, the observed currents will distribute themselves over a transition plot typified by the broken curve which intersects in turn the several true Richardson lines, shown dotted, for  $\theta_w = 0.1, 0.2$ , etc., in the area between the lines  $bd$  ( $\theta_w = 0$ ) and  $bc$  ( $\theta_w = 1$ ). The course of this transition curve depends upon the rates of adsorption, evaporation, oxidation, etc., which determine the rate of passage from  $\theta = n$  to  $\theta = m$ , where  $n$  and  $m$  are any fractions. So long as it is impossible to make  $\theta$  equal to 1 to a higher accuracy than that which, in view of the rapid volatilization of oxide, is available in practice, a nearly linear

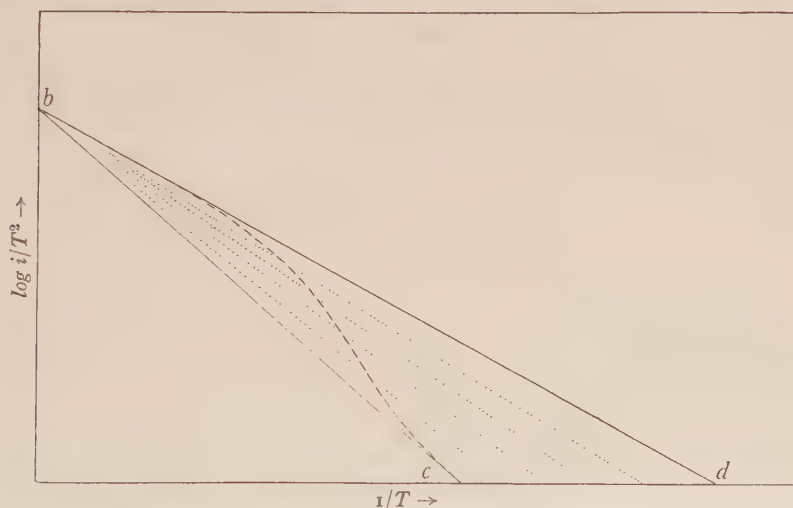


Figure 1. Transition curve between Richardson plots.

portion of an observed plot does not yield any true  $\phi$  and  $A$  but only apparent constants; these may be considerably in excess of even the true values for the fully oxygenated metal, being merely tangents to some portion of the transition curve. By a series of measurements of pressure and temperature we have proved in detail that the data usually quoted from the work of Kingdon belong to this category of apparent  $\phi$  and  $A$ .

To determine a set of values of  $\theta_w$ , we draw on a large scale a diagram such as that typified in figure 1, inserting all our experimental curves. First the line corresponding to  $bd$  is obtained from our measurements in the highest vacuum and agrees with the standard determinations of  $A_{H^+}$  and  $\phi_{H^+}$ . Secondly, that corresponding to  $bc$  is fixed by making a line whose slope gives  $\phi_{OH^+} = 6.35$  meet  $bd$  at  $1/T = 0$ , thus fulfilling the two consequences of accepting Reimann's contact potentials and the magnitudes of observed emissions.

It is to be noticed that temperature-dependence of any true work function is essentially a second-order correction as compared with the difference between these



apparent and true functions, and is neglected throughout, so that all true Richardson plots are taken as straight and only the transition plots as curved.

The diagram fulfils the conditions under which our equation (5) is equivalent to equation (4), so the Richardson plots for various values of  $\theta_w$  between 0 and 1 will lie between  $bc$  and  $bd$ , as dotted in figure 1. These are inserted in the large working graph by geometrical construction, slopes for  $\theta_w = 0.1, 0.2$ , etc. being obtained from the slopes for the work functions  $\phi_{OW} = 6.35$  and  $\phi_W = 4.64$  by the relationship of equation (4). For any observed current and temperature,  $\log i/T^2$  and  $1/T$  then constitute a pair of coordinates determining some particular  $\theta_w$  as they meet on some particular line intermediate between  $bd$  and  $bc$ .

Any other mode of deducing  $\phi_{OW}$  would of course expand or contract the scale of the  $\theta_w$  thus obtained, but it is clear that no such amendment from thermal measurements is admissible, since the latter only yield a transition plot; in particular, Kingdon's  $A$  and  $\phi$  would make  $bc$  and  $bd$  intersect at a finite temperature, giving larger emission from gas-covered than from clean metal, which is contrary to the nature of an electronegative layer.

#### § 4. THE TEMPERATURE VARIATION OF $\theta/(1-\theta)$ FOR EACH FORM OF $\theta_x$

By the ordinary arguments in the physical chemistry of reactions and the physics of change of state, a plot of  $\log_e \{\theta/(1-\theta)\}$  against  $1/T$  represents in its slope the energy exchanged in the processes which contribute to growth of  $\theta$ . Since this is only true in so far as  $\theta$  measures actual values of  $N/N_1$  or the fractional concentration of deposit, and since we have shown that neither  $\theta_a$  nor  $\theta_w$  can follow  $N/N_1$  accurately over their whole range, the energy is only reliably obtained if inferences from each approximation in its own appropriate portion of the range agree with inferences from other experiments. Accordingly we plot in figure 2 this temperature dependence of  $\theta_a$  determined by inserting the data in equation (3), and also the temperature dependence of  $\theta_w$  obtained from the same data by the method of evaluating  $\theta_w$  explained in the preceding section; in each instance three typical sets of measurements are used, curves 15, 19, 17, corresponding to highest, medium, and lowest gas pressures respectively, all at oxygen pressures between  $10^{-3}$  and  $10^{-5}$  mm.

From our physical theory of  $\theta_a$  and  $\theta_w$ , it is to be expected that the course of the former will tend more nearly to agree with that of  $\theta$  itself at the lowest pressures and highest temperatures, i.e. at the smallest values of  $\theta$ , since a very sparsely separated distribution of adsorbed particles will be unable to affect the large uncovered areas between; but as  $\theta$  grows the tendency will be for  $\theta_w$  to represent the molecular state of surface more nearly, as the whole area will become affected by overlapping fields. Hence the portions of these experimental graphs likely to afford the most reliable information as to the temperature dependence of  $\theta$  or  $N/N_1$  are  $AB$  and  $CD$ , where high temperature and low pressure support the approximation  $\theta_a$  and low temperature and high pressure support the approximation  $\theta_w$  respectively. Owing to this way in which the temperature and pressure variables



combine there is no advantage in attempting to join  $\theta_d$  and  $\theta_w$  into a single curve for any given pressure, and we only regard as significant the maximum slope of the one and the minimum slope of the other, since that is where their respective underlying assumptions tend to be fulfilled; numerical comparison between values of  $\theta_d$  and of any  $\theta_w$  obtained by any method of estimating  $i_1$  shows that for given pressure and temperature the two approximations to  $\theta$  may differ widely in absolute magnitude. But since  $\theta$  is of importance in its temperature coefficient rather than its

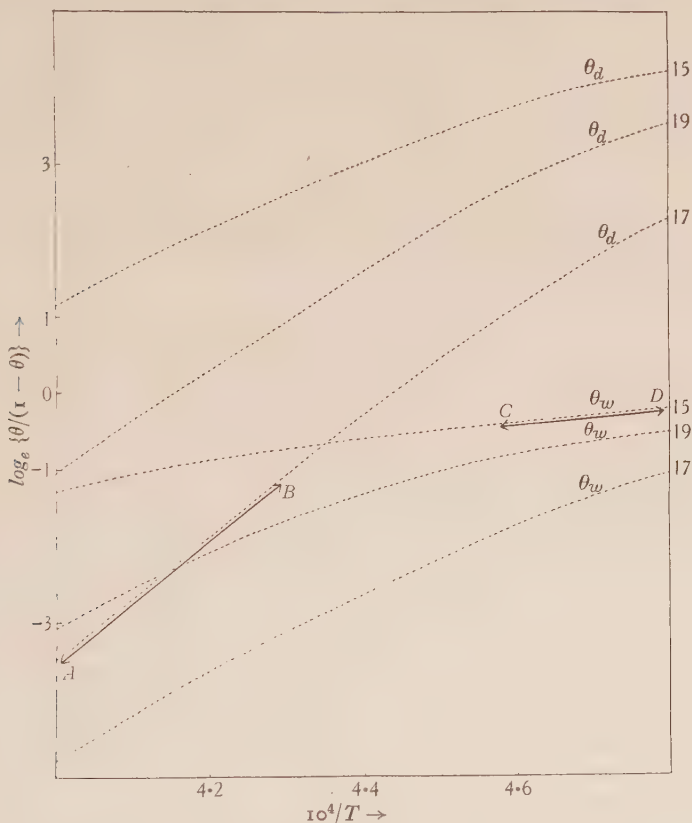


Figure 2. Temperature dependence of surface covering.

magnitude at any point, there is no need to do more than compare the slopes at *A* and *D*. This will be of interest as comparing the temperature dependence of gas-concentration in very sparse and in very dense adsorbed layers respectively, and can be related as follows to facts previously known concerning the energy needed to remove oxygen in various forms from tungsten.

##### § 5. COMPARISON WITH OTHER EXPERIMENTS

*AB* and *CD* are selected not only because each approaches the behaviour of a genuine  $\theta$ , but also because under the same two extreme sets of conditions the complex action of oxygen on tungsten approaches to the simple states commonly

distinguished as "oxygenation" and "oxidation" respectively; the slopes intermediate between *AB* and *CD* represent not merely the failure of  $\theta_a$  and  $\theta_w$  to follow  $\theta$  from zero to unity, but also the merging of the temperature dependence of adsorption at small  $\theta$  into the different temperature dependence of chemical reaction at large  $\theta$ . For this general reason the graphs would exhibit curvature even if  $\theta$  were correctly obtainable over the whole range.

Since this chemical reaction at the surface results in slow disappearance of pure metal and pure gas, a tungsten-oxygen interface is not completely described by reversible processes. Hence, although the repeatability of our observations and their independence of the order of sequence indicates that every value of  $\theta$  and of pressure can be steadied during the time of each experiment, nevertheless no simple balance can represent the complete relation between  $\theta$ ,  $T$  and pressure  $p$  or number  $\nu$  of gas impacts per  $\text{cm}^2$  per sec. Accordingly any isotherm of the Langmuir-Frenkel type suited to any small range of  $\theta$  will merge into another with a different temperature coefficient in any adjacent range. Let any such restricted equilibrium be expressed by Langmuir's relation

$$\theta = \frac{\alpha\nu}{1 + \alpha\nu},$$

$$\frac{\theta}{1 - \theta} = \alpha\nu.$$

Let a temperature dependence be added,

$$\alpha = Ce^{E/\kappa T};$$

$E$  calories per mol. represents the energy required for any action resulting in loss of oxygen from the surface. This was first introduced by Frenkel, but the generality of its present definition precludes our identifying  $C$  with any particular time interval such as that originally postulated by Frenkel.

This generalization of the well-known grounds for obtaining  $E$  from the slope of  $\log \{\theta/(1 - \theta)\}$  against  $1/T$  will then refer only to that small region over which the restricted equilibrium holds and there is reason to suppose that a fixed  $E$  is applicable; outside each such region  $E$  will be modified and possibly also the function of  $\nu$  as in the different power laws of Fowler and of Roberts.

Two such quantities  $E$ , controlling the loss of oxygen from tungsten, are known from measurement, although different authors give different molecular and atomic explanations of them. (a) At the lowest gas pressures oxygen is always found to be very strongly attached to the pure metal and slow to evaporate, the  $E$  being given by Langmuir and Villars<sup>(5)</sup> as 162,000 cal., by the present authors<sup>(6)</sup> as 147,000, and by Roberts<sup>(7)</sup> as of the order of 130,000. (b) At higher pressures a rapid oxidation is always found to set in,  $WO_3$  evaporating readily as fast as it is formed: using some experiments of Langmuir, Rideal and Wansbrough-Jones<sup>(8)</sup> have estimated the energy required for activation to this more volatile state as 20,000 cal., and on different assumptions we have used the same data to obtain 25,000. Hence the mode of escape of oxygen from tungsten, in opposition to the growth of  $\theta$ , ranges from the evaporation of an oxygenation layer requiring about 150,000 cal. for its

removal, to the instant removal of an oxide layer requiring about 20,000 only for its formation, each of these  $E$ 's being experimentally defined by an observed temperature coefficient. Hence the slope of our graph of  $\log \{\theta/(1-\theta)\}$  plotted against  $1/T$  should reach approximately 150,000 at  $\theta \rightarrow 0$  and  $\nu \rightarrow 0$  and 20,000 when  $\theta$  and  $\nu$  become large. These are the two regions  $AB$  and  $CD$  respectively in figure 2, where  $\theta_a$  and  $\theta_w$  each in turn attains some validity as an approximation to the behaviour of  $\theta$ .

Measurement of figure 2 yields 160,000 and 20,000 as limiting slopes under these two conditions, suggesting that although these thermionic methods may give absolute values of  $\theta$  much in excess or defect of actual gas concentration, their temperature variation is correct within the accuracy of experiment in those regions where each approximation in turn becomes valid.

#### § 6. CONCLUSION

A set of thermionic currents from tungsten, measured in the presence of very small and approximately constant traces of oxygen, can be analysed in two ways for obtaining information as to  $\theta$ , the fraction of surface covered. Of the current-deficiency approximation  $\theta_a$  and the work-function approximation  $\theta_w$  the former is more capable of expressing a correct course of the way surface conditions vary at high temperature and low gas pressure, and the latter at low temperature and high gas pressure. After devising a new method for obtaining the work functions of a partially oxygenated surface, we have plotted a graph of  $\log \{\theta/(1-\theta)\}$  against  $1/T$  for three gas pressures according to each of the two approximations  $\theta_a$  and  $\theta_w$ . Measurement of the low-temperature, high-pressure and high-temperature, low-pressure extremes of the graphs yields temperature coefficients corresponding to 160,000 cal. and 20,000 cal. respectively, from the slopes to which they tend where the above conditions for the validity of each approximation in turn is fulfilled. These values agree with the high energy of oxygen evaporation and the low energy of oxidation, previously estimated as from 130,000 to 162,000 and 20,000 or 25,000 respectively.

#### § 7. ACKNOWLEDGEMENT

We are very grateful to Prof. S. W. J. Smith, F.R.S. for his kindly provision of all facilities for this work.

#### NOTE ADDED 15 MARCH 1937

When the concentration of a deposit is such as to justify the common term "film", however dilute,  $\theta_w$  is the more reliable approximation to  $N/N_1$ . But when the adsorbed atoms are so few and widely separated that the regions not contributing to clean emission are isolated,  $\theta_a$  is the more reliable, as it is not possible to average the work function over such a discontinuous surface.

Since there is no direct evidence as to the concentration at which  $\theta_w$  supersedes  $\theta_a$  for the case of  $OW$ , our curves are not claimed as independent data for determining  $E$ . If  $\theta_w$  were valid throughout all concentrations, our slope would give the

$E$  of evaporation as 110,000 and the  $E$  of oxidation as 20,000, while if  $\theta_a$  were valid throughout the slope would give 160,000 and 35,000. But as there is independent evidence that these quantities are not far from 150,000 and 20,000 respectively, our results may be taken as indicating that the temperature coefficient of  $\theta_a$  is a fair approximation under just those conditions that give 160,000, while the temperature coefficient of  $\theta_w$  is preferable under the conditions that give 20,000.

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## DISCUSSION

L. R. G. TRELOAR. The paper gives no information concerning a problem of very considerable physical interest, namely, the way in which the work function varies with the actual number of oxygen atoms present on a tungsten surface, i.e. with the true value of  $\theta$ . Equation (4) defines a surface covering  $\theta_w$  such that the relation between  $\phi$  and  $\theta_w$  is linear. Now in the case of electropositive contaminations, for which the true relation between  $\phi$  and  $\theta$  is known, it does not even approximate to linearity. For the case of oxygenated tungsten it seems not unlikely that there may be a similarly wide departure from linearity. If this is so, it seems at first sight rather strange that the application of equation (4), even over a small range of  $\theta$ , should lead to the correct value of the energy associated with the oxidation process.

AUTHORS' reply. The solution of Mr Treloar's problem would constitute a great advance in the subject, but the title of our paper implies correctly that it lies outside our present purpose. We have explained in § 2 that this is because no reliable method yet exists for counting the oxygen particles on the surface as has been done for alkalis: electropositive experiments can be carried out far below the condensation temperature of the alkali vapours involved, but for oxygen the latter is less accessible. Thus even a molecular-ray method cannot decide with precision how many particles have reached the filament and how many remain there. Hence at present the only access to  $\theta$  with oxygen is through these approximations, for whose validity the correct value of the energy-slope affords some support. Actually, since the departure from linearity of  $\phi/\theta$  for alkalis is due to cohesion between the adsorbed particles, this is not to be expected to the same extent with oxygen, which binds more to the tungsten and less to itself. At larger values of  $\theta$  than we ourselves use, other breakdowns of the relation occur which, however, do not concern the subject-matter of this paper.



# THE THERMAL AND ELECTRICAL CONDUCTIVITIES OF A SAMPLE OF ACHESON GRAPHITE FROM 0° C. TO 800° C.

BY R. W. POWELL, B.Sc., Physics Department, National Physical Laboratory, Teddington

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**ABSTRACT.** Determinations have been made of both the thermal and the electrical conductivities of a sample of Acheson graphite over the temperature-range from 0° C. to 800° C. The thermal conductivity  $K$  is found to decrease from a value of 0.40 unit at 0° C. to 0.16 at 800° C., the rate of decrease being greater at lower temperatures. The electrical conductivity  $\sigma$  has at 0° C. a value of  $1218 \Omega^{-1}\text{-cm}^{-1}$ , which increases to a maximum value of 1518 at 450° C. and decreases to 1444 at 800° C. The corresponding Lorenz function  $K\sigma^{-1}T^{-1}$  has a value of  $121 \times 10^{-8}$  at 0° C., which is more than 200 times the value indicated by the electronic theory and possessed by most well-conducting metals. With increase in temperature the value of the Lorenz function decreases rapidly, its value being given by the equation  $K\sigma^{-1}T^{-1} = 0.0294 T^{-1.8}$ . Thus the Wiedemann-Franz-Lorenz law does not hold for the present sample of graphite, but is replaced by the equation  $K\sigma^{-1}T^{0.8} = \text{a constant}$ . The power to which the absolute temperature is raised is different, as well as the constant value of the expression.

## § 1. INTRODUCTION

IN an earlier paper<sup>(1)</sup> values have been published for both the thermal conductivity  $K$  and electrical conductivity  $\sigma$  of several heat-resistant alloys. Some of these values showed considerable departures from the Wiedemann-Franz-Lorenz law, according to which  $K\sigma^{-1}T^{-1}$  is constant. The greatest departure occurred at 0° C. where, for two of the metals tested, the value of the Lorenz function was about double that indicated by the electronic theory and possessed by most good conductors. The extent of the deviation decreased with increase in temperature.

Graphite has a thermal conductivity of the same order as that of many metals, yet according to Koenigsberger and Weiss<sup>(2)</sup>, Barratt<sup>(3)</sup> and Störmer<sup>(4)</sup>, its Lorenz function in the neighbourhood of room-temperature is some 200 to 400 times as great as the normal value. It was considered of interest to ascertain whether the extent of this deviation would also decrease with increase in temperature, and for this purpose determinations of the thermal and electrical conductivities have now been made on a sample of Acheson graphite over a range of temperature similar to that covered by the earlier experiments.

## § 2. EXPERIMENTS

The graphite selected for use in this investigation was a sample of Acheson graphite which was originally in the form of a rod some 9 cm. in diameter and 75 cm. long. From this sample a specimen 7.34 cm. in diameter and 38 cm. long was prepared. This was first tested over the temperature-range from 30° C. to 200° C. by a method similar to that already described for the heat-resistant alloys<sup>(1)</sup>.

The rod was mounted vertically within a coaxial cylindrical guard tube, and the interspace was packed with heat-insulating powder. The lower end of the rod was electrically heated, and the temperature-gradient established in the rod in the final matched steady state was measured by means of thermocouples. The heat flowing along the rod was measured by means of a water-flow calorimeter attached to the upper end, the thermal contact between the graphite and the calorimeter being improved by means of a small mercury bath.

The rod of graphite was later turned down to a smaller diameter of 3.85 cm. and the thermal-conductivity measurements were repeated. For this smaller size of rod a water-flow calorimeter was used which consisted of a coil of tubing soldered to a slightly tapered brass tube about 10 cm. in length and 3.8 cm. in maximum diameter. This unit made good contact with a similar taper on one end of the graphite rod. The values of the thermal conductivity obtained for both sizes of rod are plotted in figure 2, and it will be seen that the two sets of data are in close agreement. The specific resistances also of the two rods agreed. These were measured by the usual method in which the fall in potential along a length of the rod was compared with that across a shunt of known resistance when the same current was passed through each.

To extend the measurements to higher temperatures it was necessary to use an evacuated enclosure to prevent oxidation of the specimen. For these tests the rod 3.85 cm. in diameter was further machined to have a diameter of 1.47 cm. A 20-cm. length of this thin rod was first tested in an apparatus which formed part of an exhibit at the Physical Society's twenty-fifth annual exhibition of scientific instruments and apparatus, and which was subsequently illustrated and described in a published account of the exhibition<sup>(5)</sup>.

The energy input to a platinum heating coil wound on a steatite former and enclosed in an axial hole in the top of the graphite rod was measured. Lateral loss of heat was prevented by the usual matched-guard-tube method, and loss in an upward direction was prevented by mounting a guard heater above the top of the rod and guard tube, and adjusting its energy until a thermocouple located between this heater and the top of the specimen indicated the same temperature as did thermocouples pegged into the upper parts of the rod and guard tube. The current and potential leads and the thermocouple wires were all brought out at the base, vacuum-tight seals being made by water-cooled wax-filled vessels.

In this set-up results were obtained for both thermal and electrical conductivities up to a temperature of about 800° C. It will be seen from figure 2 that over the temperature-range from 200° C. to 300° C., which was common to both the

high-temperature and the low-temperature determinations, the values agree to within 2 or 3 per cent. At these temperatures the method employed for the elimination of heat-losses from the small rod would appear to be satisfactory. For tests at higher temperatures it was necessary to verify that the observed energy-input gave a true value for the flow of heat in the specimen. It was found impracticable to do this by measuring the temperature-gradient at the cold end of the rod, and

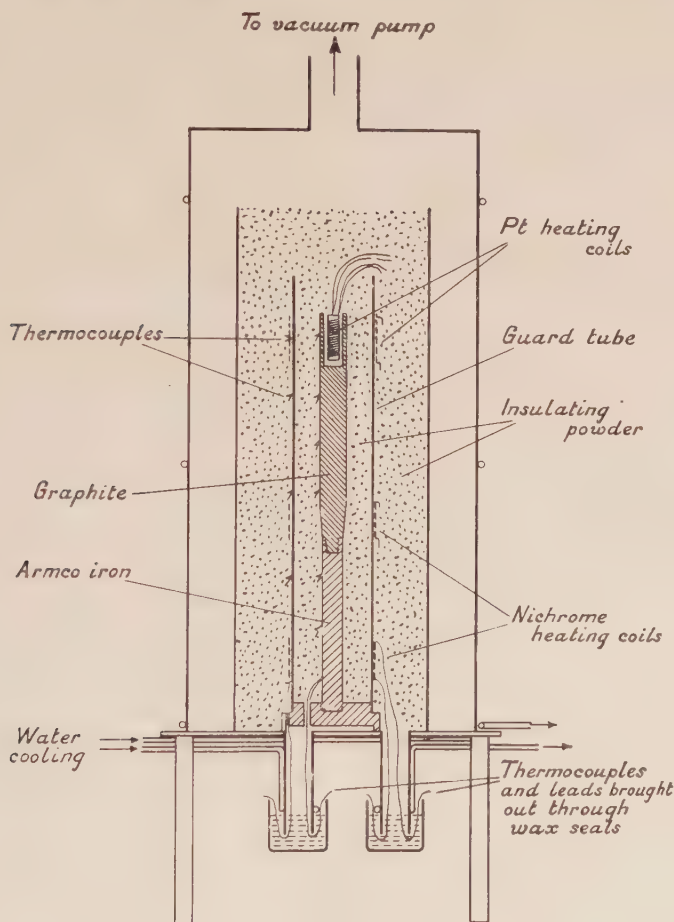


Figure 1. Diagram of apparatus.

a method resembling that used for the heat-resisting alloys<sup>(1)</sup> was eventually adopted, a short rod of the graphite being screwed into a rod of Armco iron of known conductivity<sup>(6)</sup>. The details of the present set-up in the evacuated enclosure are shown in figure 1.

The values obtained for the thermal conductivity when this method was used are indicated in figure 2 and are seen to be in general agreement with those obtained in the previous experiment, except that they tend to lie on a somewhat lower curve at the higher temperatures. In drawing the final curve through the experimental

points somewhat greater reliance has been placed on these latter values. With neither set-up was it found possible to extend the tests to temperatures much above  $800^{\circ}\text{C.}$ , owing to failure of the upper heating coils.

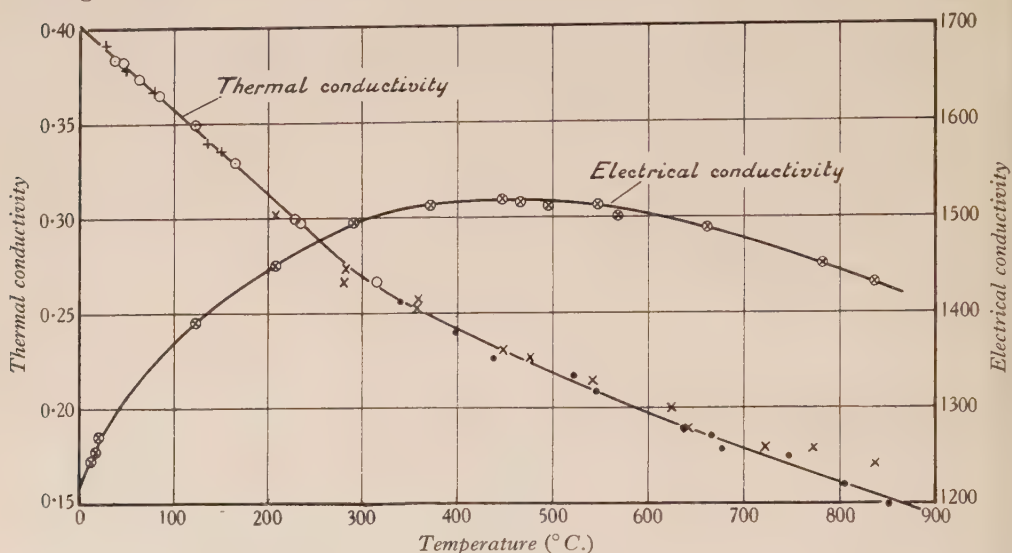


Figure 2. Variation of conductivities of Acheson graphite with temperature. Measured on thick rod,  $\odot \odot \odot$ ; on medium rod  $+++$ ; on thin rod by first method  $\times \times \times$ ; on thin rod by second method  $\dots$

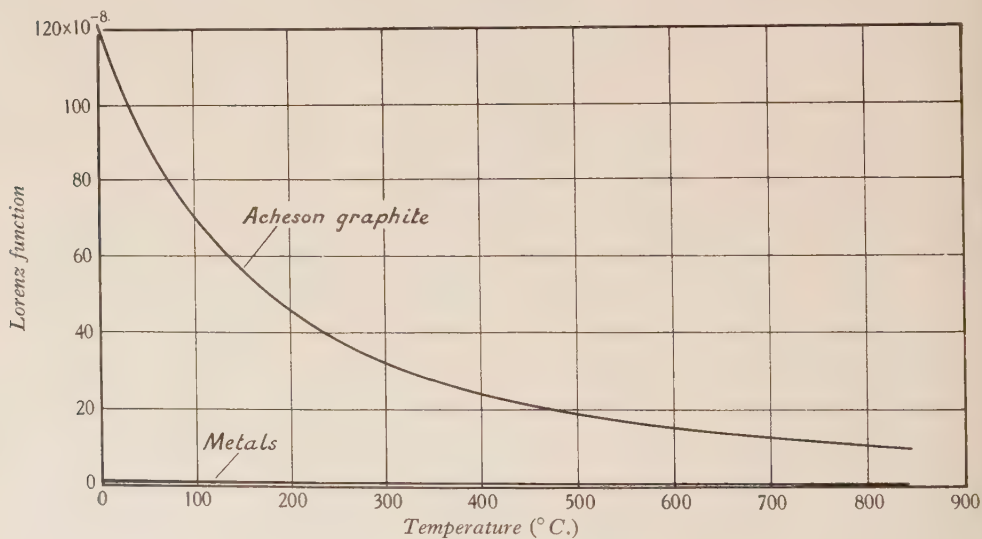


Figure 3. Variation of Lorenz function with temperature.

The curve showing the variation of electrical conductivity with temperature also is plotted in figure 2, whilst that for the Lorenz function, which has been derived from the smoothed values read from the thermal and electrical conductivity curves, is drawn in figure 3. For the purpose of comparison a curve giving the Lorenz function of normal metallic conductors has been included in the same figure.



### § 3. DISCUSSION OF RESULTS

*Thermal and electrical conductivities.* Graphite is produced when carbon is heated to extremely high temperatures. The physical properties of graphite differ considerably from those of carbon and vary to a lesser extent from sample to sample. The variation presumably depends on the completeness of the transformation, and thus on the temperature of firing, as well as on the purity of the material.

Published values for the thermal conductivity of graphite in the neighbourhood of atmospheric temperature have ranged from 0.037 to 0.85 cal. per cm. per sec. per degree, the corresponding values of the electrical conductivity being 52 and 3000  $\Omega^{-1}\text{-cm}^{-1}$ . The results for the more poorly conducting graphite were due to Barratt, the others to Koenigsberger and Weiss. Barratt's material, which was a lead extracted from a pencil, would seem to be an unusually poor graphite. Its electrical conductivity was less than that of amorphous carbon, which according to Störmer has an electrical conductivity of 114  $\Omega^{-1}\text{-cm}^{-1}$  and a thermal conductivity of 0.0048 unit at 13° C.

The present results fall between the above extreme values, and it should be realized that they apply to a particular sample of Acheson graphite. In a future paper the author hopes to deal with the thermal and electrical conductivities of a number of varieties of graphite.

It will be seen from figure 2 that whereas the thermal conductivity of Acheson graphite decreases with increase in temperature throughout the temperature-range covered by the experiments, the electrical conductivity increases to a maximum value at a temperature of about 450° C., and then decreases with further increase in temperature. This maximum value is 1.25 times the value indicated at 0° C. by extrapolation of the curve. The nature of the curve showing the variation of electrical conductivity with temperature agrees with that found for graphite by several other workers, including Noyes<sup>(7)</sup> and Nishiyama<sup>(8)</sup> but the temperature at which the maximum value occurs differs from sample to sample. The former used a rod of Acheson graphite for which he found a maximum value of 1.29 times the value at 0° C. to occur at a temperature of about 550° C. Nishiyama studied carbon filaments which had been graphitized to increasingly high amounts by heating to successively higher temperatures. It appears from his results that as the extent of graphitization is increased the maximum occurs at lower temperatures, and at the same time the ratio of its value to that at 0° C. decreases. An X-ray analysis of his specimens was undertaken by Nishiyama, and he considered that the electrical-conductivity results could be explained by the increased grain-size which accompanied increased graphitization.

Three determinations of the thermal conductivity of graphite have been made by other workers over the temperature-range covered by the present experiments. Hansen<sup>(9)</sup> determined the thermal conductivity of a 1-inch rod of Acheson graphite electrically heated at one end and water-cooled at the other. The rod was lagged, but no guards were used, so that the heat supplied exceeded that collected by the water-flow calorimeter. According to the values derived from the latter measure-

ments, which are probably the more correct, the thermal conductivity decreased from 0.45 at 100° C. to 0.23 at 600° C. and lay from 25 to 15 per cent above that of the present sample of graphite. Hering<sup>(10)</sup> derived values for the thermal conductivity of graphite from observations on furnace electrodes. Mean values over large temperature-differences are obtained, and these values are probably less exact. They are, however, of the same order of magnitude and show a decrease from 0.34 at 245° C. to 0.29 at 510° C. Crary<sup>(11)</sup> has more recently carried out a determination on Acheson graphite, the source of heat being a flat coil sandwiched between two similar cylinders of graphite 4 in. in diameter. His values for the thermal conductivity range from 0.42 at 0° C. to 0.20 at 730° C. At these extreme temperatures the thermal conductivity is thus only 5 and 15 per cent greater than that of the present experiments. Crary's {conductivity, temperature} curve differs, however, in that it is convex with regard to the axis, and at 400° C. the value of the thermal conductivity is 0.34 compared with the 0.24 obtained in the present investigation. The explanation of this difference is not apparent, but it might be remarked that the method adopted by Crary for the prevention of lateral heat-loss appears to be unsatisfactory in that it assumes the material under test to have a constant thermal conductivity, whereas the conductivity of graphite decreases appreciably with increase in temperature.

*The Lorenz function.* It will be seen from figure 3 that the Lorenz function of Acheson graphite at 0° C. is some 200 times as great as that of most metals. With increase in temperature its value decreases rapidly, and graphite thus appears to fall in line with such metals as bismuth, nichrome, and F.H. stainless steel in that its Lorenz function approaches the electronic value when the temperature becomes sufficiently high.

A comparison of columns 4 and 5 in table 1 shows that the variation of the Lorenz function with temperature can be represented by the equation

$$L = K\sigma^{-1}T^{-1} = 0.0294T^{-1.8} \quad \text{.....(1),}$$

$$\text{or} \quad K\sigma^{-1}T^{0.8} = 0.0294 \quad \text{.....(2),}$$

where  $L$  is the Lorenz function,  $K$  the thermal conductivity in cal. per cm. per sec. per degree,  $\sigma$  the electrical conductivity ( $\Omega^{-1}\text{-cm}^{-1}$ ), and  $T$  the temperature (° K.).

Table 1. Comparison of values of  $L$  derived experimentally with those given by equation (1)

$T$	$K$	$\sigma$	$L$ as $K\sigma^{-1}T^{-1}$	$L$ as $0.0294T^{-1.8}$
273	0.402	1218	$120.8 \times 10^{-8}$	$121.1 \times 10^{-8}$
373	0.357	1369	70.0	69.1
473	0.312	1445	45.7	45.0
573	0.268	1497	31.3	31.9
673	0.240	1515	23.5	23.9
773	0.217	1517	18.5	18.6
873	0.197	1503	15.0	14.9
973	0.178	1476	12.4	12.3
1073	0.160	1444	10.3	10.3

Extrapolation indicates that at 3800° K., which approximates to the arc-temperature, the Lorenz function of graphite is about  $1 \times 10^{-8}$ , which is still somewhat greater than the theoretical value for metallic conductors.

This investigation has shown that although graphite has a thermal conductivity of the same order as that of many metals, it differs in that the Wiedemann-Franz-Lorenz law is not even approximately true. For the present sample of graphite this law can be replaced by the equation  $K\sigma^{-1}T^{0.8} = \text{a constant}$ , in which changes have been made in both the power to which the absolute temperature is raised and the constant value of the expression.

#### § 4. ACKNOWLEDGEMENTS

The author desires to thank Dr G. W. C. Kaye, Superintendent of the Physics Department, for his continued interest in this investigation and for the provision of the necessary facilities. He is also indebted to Mr E. E. Smith, B.Sc. for assistance in the preparation of the specimens used in the earlier stages of the investigation and for taking some of the observations, and to Mr M. J. Hickman for rendering similar assistance in the final experiment at high temperatures.

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#### DISCUSSION

Prof. G. I. FINCH. Structurally graphite consists of parallel hexagonal-network sheets of carbon atoms. The bindings between the carbon atoms in each sheet are homopolar and very strong but the bindings from sheet to sheet are very loose, of the free electron type, and this is the cause of the colour and good conductivity of graphite. The structure also accounts for the ease with which graphite can be cleaved into thin sheets, and in our laboratory we have succeeded in cleaving off sheets comprising as few as three or four hexagon layers. Such thin sheets are, as one might expect, almost as transparent as glass, and colourless. Owing to the loose binding between the sheets, the graphite structure can be expanded like a concertina by introducing oxygen or alkali metals which do not loosen the homopolar bindings but



go in between the sheets. Thus we see that the properties of the graphite crystal in directions parallel and normal to the sheets are quite different, and we should expect to find great differences in the conductivity in a single crystal of graphite according to the direction in which the current flows. It would be of great interest to study the variation of conductivity with direction in a single crystal of graphite. The Natural History Museum possesses some very beautiful specimens containing single crystals measuring as much as 1 cm.<sup>3</sup>

MISS M. D. WALLER. I should be interested to know how the conductivities of Acheson graphite vary at temperatures below 0° C., as I have found\* maximal values of the vibration frequency or Young's modulus of specimens of Acheson graphite and carbon at about -50° C.

DR O. KANTOROWICZ. A high value of the Lorenz factor for a body like Acheson graphite does not necessarily indicate that the graphite crystals which constitute Acheson graphite show any anomaly at all. Acheson graphite is a kind of ceramic body and contains many pores, and also impurities, which must be suspected of being comparatively bad conductors for electricity and are most likely located on the grain boundaries. Consequently the electric current used to measure the resistance of the sample will flow from grain to grain through those rather restricted passages where one graphite crystal actually touches its neighbouring crystals, and the resistance to electricity of such an agglomerate is rather high. On the other hand the heat-flow is to a far lesser degree affected by the pores and impurities, both of which possess a certain conductivity for heat. The heat therefore flows along a path different from that of the electric current, and partly through other substances. Under these circumstances the quotient of electric and thermal conductivity is probably more a measure for the porosity and impurity of the particular sample than a characteristic of the constituent graphite crystals.

AUTHOR'S reply. I agree with Prof. Finch that it would be interesting to study the conductivities of a single crystal of graphite and appreciate the suggestion as to a possible source from which a suitable crystal might be obtained.

The experiments quoted by Miss Waller are of particular interest, as her observations on the elastic constants presumably indicate some change in the crystalline properties of graphite, and one inference from the high Lorenz function of this material is that a large proportion of the heat is conducted by crystalline rather than electronic processes. So far as I am aware the thermal conductivity of graphite has not been studied at temperatures below 0° C. and I hope to extend my measurements on graphite to lower temperatures.

The factors mentioned by Dr Kantorowicz may also play some part in explaining the high Lorenz function of graphite. X-ray analysis has not revealed any appreciable percentage of crystalline impurity in the specimen used in this work but amorphous carbon is probably present. It is hoped that the tests now in progress on other specimens of graphite will throw further light on the problem.

\* *Proc. roy. Soc. A*, 157, 388 (1936).

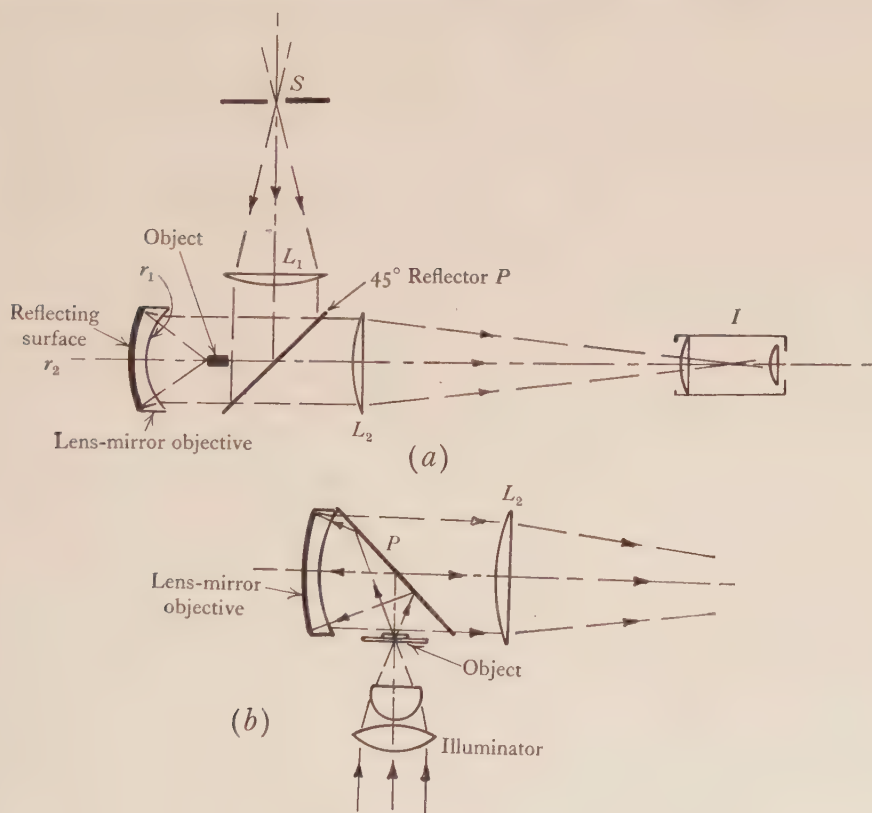


## DEMONSTRATION

A MICROSCOPE FOR USE WITH ULTRAVIOLET LIGHT, EMPLOYING A REFLECTING OPTICAL SYSTEM OF APLANATIC PROPERTIES.

*Demonstration given on 28 May 1937 by B. K. JOHNSON*

Experiments have shown that transparent and opaque objects show marked differential absorption and reflection effects when observed with ultraviolet illumination under the microscope. By choosing appropriate wave-lengths, the desired



*Reproduced by permission of the Journal of Scientific Instruments*

The lens-mirror system as applied to the examination of (a) opaque objects, and (b) transparent objects.

amount of contrast in the image can be obtained; this act suggests a possible means of eliminating staining processes. In order to examine a specimen over a considerable range of ultraviolet wave-lengths it is necessary to have a number of quartz objectives, for it is not yet possible to achromatize the latter. This fact has led the author to design a simplified microscope objective in the form of a lens-mirror and

one single lens. Up to numerical apertures of 0.55 such a system can be made free from spherical aberration and coma, whereas all surface-reflecting systems (such as the ellipsoid, the parabola, etc.) suffer from large amounts of coma immediately off the axis. The design of this optical system lends itself for use in the visible region of the spectrum provided that monochromatic light be used, and also in the infra-red. Its chief advantages over the orthodox quartz refracting objectives are (1) simplicity of manufacture and increased accuracy in the making of the surfaces; (2) increased contrast in the image due to reduction of number of air-quartz surfaces; (3) elimination of back-reflections when opaque objects are examined.

A full description of the instrument has been published.\*

\* *J. Sci. Instr.* **11**, 384 (1934).

## REVIEWS

*The March of Science, 1931-1935.* By various Authors. Pp. viii + 215. (London: Pitman and Sons, 1937.) 3s. 6d.

After a long period during which specialization has been increasing, and has indeed been an explicitly announced goal, we seem to be entering on one with somewhat different characteristics. Specialization is of course not losing its importance, but it is now realized that the man who is only a specialist is not thereby superior to another who, in addition to his specialist knowledge, has also a background of wider culture. The changing viewpoint is to be found expressed in the frequent discussions of the examination system and the school curriculum, and is even reflected in popular literature. Wells unifies history, so that specialists in prehistory, archaeology, and the other branches are forced to meet each other and each to see his science against the background of the others. The same tendency is to be recognized in the recent publications of the masters of exposition, such as Andrade, J. A. Thomson and others.

When such a tendency is to be discerned, who better than the British Association should encourage and foster it? That Association was formed for the dissemination of knowledge, and the sections cover between them practically all science. In this book, which is issued under the authority of the Council, eminent practitioners of more than a dozen individual sciences give some account of the main lines of growth in their fields of work during the last five years. Naturally, their methods of treatment vary considerably, and they evidently differ widely as to the standard of knowledge which can be assumed in their readers, as well as in the degree to which they assume these readers desire to pursue the subject. Thus, whereas the physicist deems it desirable to offer an elementary explanation of how a probability wave can represent an electron, and to give us a rapid sketch of the growth of particle physics since 1895, the anthropologist speaks on his first page of the Icenian, Cromerian, Clactonian, Levalloisian, Mousterian, pre-Chellean, Acheulian and Micoquian cultures, and seems to suppose that they represent for us concepts as definite as those which they represent to him. Again, the zoologist gives a bibliography of more than 100 references, in four or five different languages.

On the whole, however, the authors seem to agree in a general way that the man in the street is an intelligent fellow who has some acquaintance with the achievements of science and a real desire to learn more—in short that he is the typical member (or rather, the ideal member) of the British Association.

The existence of this book, with its collection of surveys of recent advances in nearly all the sciences, naturally prompts us to ask the question, "What have been the greatest advances in science in that period?" The answer must depend a good deal on individual bias, but a survey of these chapters certainly seems to make out a good case for psychology as the recipient of the prize for rate of progress. Economics, it is true, may have changed more (and more often) than the others, but the result bears no evident signs of being a better science than its predecessors, whereas all the others do bear the outward signs of an approach to truth.

J. H. A.

*Structure of Atomic Nuclei and Nuclear Transformations*, by G. GAMOV. Pp. xii + 270. (Humphrey Milford, Oxford University Press, 1937.) 17s. 6d. net.

Though formally a second edition of the author's well-known *Constitution of Atomic Nuclei*, the present volume is to all intents and purposes a new work. The astounding developments which have taken place in the last five years (including on the experimental

side the discovery of the neutron and the production of artificial radioactivity, and on the theoretical side the fuller realization of the importance of interaction forces) have both revolutionized nuclear theory and widened its field. Very little of the old book survives in the present pages. It would be interesting, but fruitless, to speculate as to the contents of the next edition. Enough to say that in the present edition the argument moves freely and easily in a region so highly congested that the particles which inhabit it find it impossible to maintain even their identity.

Although the book deals with the theoretical rather than the experimental aspect of the subject, it is not unduly loaded with mathematical calculations, and no reader should be deterred from attempting it through lack of interest in the technique of wave mechanics. The author's main aim has been to discuss, in the clearest possible way, the principles employed in attempting to deduce the laws of physics within the nucleus, and the application of these laws to the problems of nuclear structure and nuclear behaviour. The interdependence of theory and experiment, the nature of the assumptions employed, and the occasional discordance between calculated and experimental results are displayed with clarity and candour. As a result, the book is not only an admirable survey of the present state of the subject, but a fascinating study of new knowledge in the making. The physicist who wishes for an adequate account of the philosophy underlying this new branch of physics will find his requirements well satisfied; and for those who may wish to pursue the mathematical calculations with greater rigour there are numerous references to original papers.

J. A. C.

*The Physics of Electron Tubes*, second edition, by L. R. KOLLER, Ph.D. Pp. xvii + 234. (International Series in Physics. London: McGraw Hill Publishing Co., Ltd.) 18s. 0d.

This book gives a very good survey of what takes place inside electronic devices in general. It does not discuss the details of circuits and methods of application of the various electronic tubes. The first four chapters deal with thermionic emission in principle, and cover thoriated tungsten cathodes, caesium on tungsten, caesium on oxygen on tungsten, and oxide-coated cathodes, and also a fair amount of general theory of thermionic emission. Secondary-emission phenomena and their applications are then dealt with, and afterwards special types of electron tubes are described. The omission of any reference to the X-ray tube here is rather surprising, and so is the brief nature of the paragraph on multielectrode tubes. Otherwise the field covered is fairly complete. There is an excellent chapter on the determination of the temperature of the electrodes in electronic devices.

The phenomena of the discharge in gases have been very ably worked into the general scheme of electronic tubes, and there follows a more practical chapter on grid-controlled arcs. There are then chapters on photoelectricity, photoconductivity, and the photo-voltaic effect. In the latter two chapters the author has deviated from the title of electron tubes rather considerably, but they are written with the same clarity and conciseness as the rest of the book and therefore form a welcome addition to the text.

This book, which has now reached the second edition, has been brought very fully up to date, and is in touch with all the latest electronic developments. Sections have been added on electron optics, secondary emission multipliers, ignitrons, and positive-ion emission. It is an excellent book for the student of physics and electrical engineering, and will also be useful to the specialist engaged on research in other branches of physics in which these electron tubes find application.

J. A. D.



*The Low-Voltage Cathode-Ray Tube*, by G. PARR. Pp. x+177, with 76 figures. (London: Chapman and Hall.) 10s. 6d. net.

This book should make a strong appeal to a large number of readers. It is sufficiently elementary for the more advanced students of physics and engineering and also sufficiently advanced to be of use to the research worker. The first chapter, entitled "Construction and operation", contains a general introduction to the principle of the cathode-ray tube. The second chapter deals with focusing and performance and is the only chapter in which some weakness makes itself felt. The treatment of electrostatic and electromagnetic focusing in this chapter is rather too scanty, and the distortion effects, as observed on the screen, are not dealt with as fully as one would have expected. The third chapter describes the formation of Lissajou's figures. This is a good feature because it revises a subject about which knowledge is too often taken for granted. The fourth chapter deals very fully with the essentials of the linear time base, and the next chapter goes on to other types of time base. The three remaining chapters deal with the various applications of cathode-ray tubes. In the first place to radio engineering, in the second place to general industrial problems, and finally to television. The chapter dealing with the industrial application is rather speculative, but the sections describing the uses of the cathode-ray tube in radio and television engineering are clear expositions of more well-known methods. The text is concise, but difficult points are explained with considerable care and the book is well illustrated throughout. The general quality of the publication is excellent.

J. A. D.

*Atlas der Analysen-Linien der wichtigsten Elements*, by Dr F. LÖWE. Pp. iv+37, with 16 plates. (Dresden and Leipzig: Theodor Steinkopff, 1936.) RM. 10.

This is a second, and much changed, edition of the author's *Atlas der letzten Linien*, the new title being in accordance with Gerlach's nomenclature. To those engaged in spectrographic detection of the small quantities of elements in solutions, ores, alloys and other materials the work can be recommended as a saver of time and trouble. The author is the head of a section of the Zeiss works at Jena. In the first few pages are notes on apparatus and method (condensed spark between carbon electrodes on which a chloride solution has been evaporated) and on other atlases and tables (titles and references are listed). Then follow the 16 plates of selected regions of line spectra of 46 elements, and, printed on opposite pages, brief explanations and lists of wave-lengths. To the latter are added data for 11 elements not included in the plates. The book ends with a ten-page table of analysis lines between  $\lambda$  1854 and  $\lambda$  8944. A notable and commendable feature is the fewness of the lines included in this table and in the plates.

W. J.

*Ions in Solution*, by R. W. GURNEY. Pp. vi+206. (Cambridge: The University Press, 1936.) 10s. 6d.

One of the characteristics that distinguish the best conjurers from those of the second rank is the fact that they send their audiences home impressed with the ease of the performance, and filled with the conviction that they have only to snap their own fingers in the air to find pennies in their hands. Dr Gurney shares these powers. Apparently provided only with the common sense necessary to depict qualitatively the form of the potential energy curve of an electron, ion or nucleus as the case may be in the presence of another particle or assemblage of particles, and furnished with a knowledge of what a Boltzmann factor is, he can draw forth the quantitative results of the Debye-Hückel theory so clearly as to persuade the reader that he could have done it himself without the help of these pioneers.

A very big claim is made in the preface, to the effect that the methods employed in the book can be expected to lead to an advance in ionic theory commensurate with that made in atomic theory by Bohr's methods. The reviewer is thus saved the trouble of deciding whether the book is or is not epoch-making—that is already decided for him. It does seem probable, however, that the claim can conscientiously be endorsed. It marks the epoch when attention is transferred from the salt in solution to the ions themselves, just as Bohr's theory completed what Ritz had commenced—the transfer of attention from spectral lines to energy levels.

This alteration in outlook leads (or will lead when the necessary quantitative data are available) to an easy understanding of the relation between ions *in vacuo*, ions in a crystal lattice and ions in solution. It permits of simple deductions relating to solubility, mutual precipitation, and electromotive forces and particularly the electrochemical series. It is very successful in dealing with optical absorption, and therefore with the colours of solutions, and must contain adequate information from which to calculate the electrical conductivity of an arbitrary solution at any degree of concentration. This last matter, however, is one to which little attention is devoted in the book under review.

The outstanding impression which the book leaves, is one of clarity, due to the facts that it is self-contained (there is not one reference in it to any scientific paper) and that the physical principles are kept so well to the fore. There are in it two suggestions that the author contemplates writing a companion volume containing a more detailed treatment, and it is to be hoped that in due course this will in fact appear.

J. H. A.

*The Organic Chemistry of Nitrogen*, by N. V. SIDGWICK, F.R.S. New edition revised and rewritten by T. W. J. Taylor and W. Baker. Pp. xix + 590. (Oxford: Clarendon Press, and London: Humphrey Milford, 1937.) 25s. net.

The first edition of this book, published in 1910, has long been out of print and scarce, and the appearance of a second completely revised edition is welcome. The book deals with selected groups of organic compounds of nitrogen which are in themselves of considerable chemical and biochemical importance, and the treatment is very complete on the chemical side, from the point of view of both the theory and the experimental aspects. What differentiates the book from most treatises on organic chemistry is the unusually full discussion of relevant physical considerations, and at the present time, when so many organic compounds are the subject of study by physicists in various fields, the book is timely and of great interest and value. The eighteen chapters deal with the important groups of simpler organic compounds of nitrogen, and there are full references to the literature. The printing and paper are excellent and the moderate price is worthy of emphatic mention. This is a book of first-rate importance, and the new edition fully maintains the excellent features of the old. All the chemical considerations which must be known in dealing with any compounds from the point of view of physics or physical chemistry are explained, and the book is one which should be included among the works on chemistry in a physical library.

J. R. P.



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